



## Radiochemical extraction of actinides in the beach areas near by a mothballed nuclear power plant

Roviello , Valentina ; Ruberti , Daniela; De Cesare, Mario; Terrasi, Filippo; Sabbarese, Carlo; De Cesare , Nicola; Hou, Xiaolin; Roos, Per; Raffaele, Buompane

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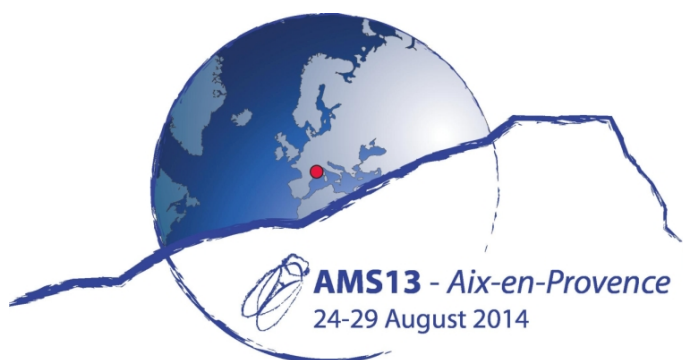
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# AMS-13

## The Thirteenth International Conference on Accelerator Mass Spectrometry

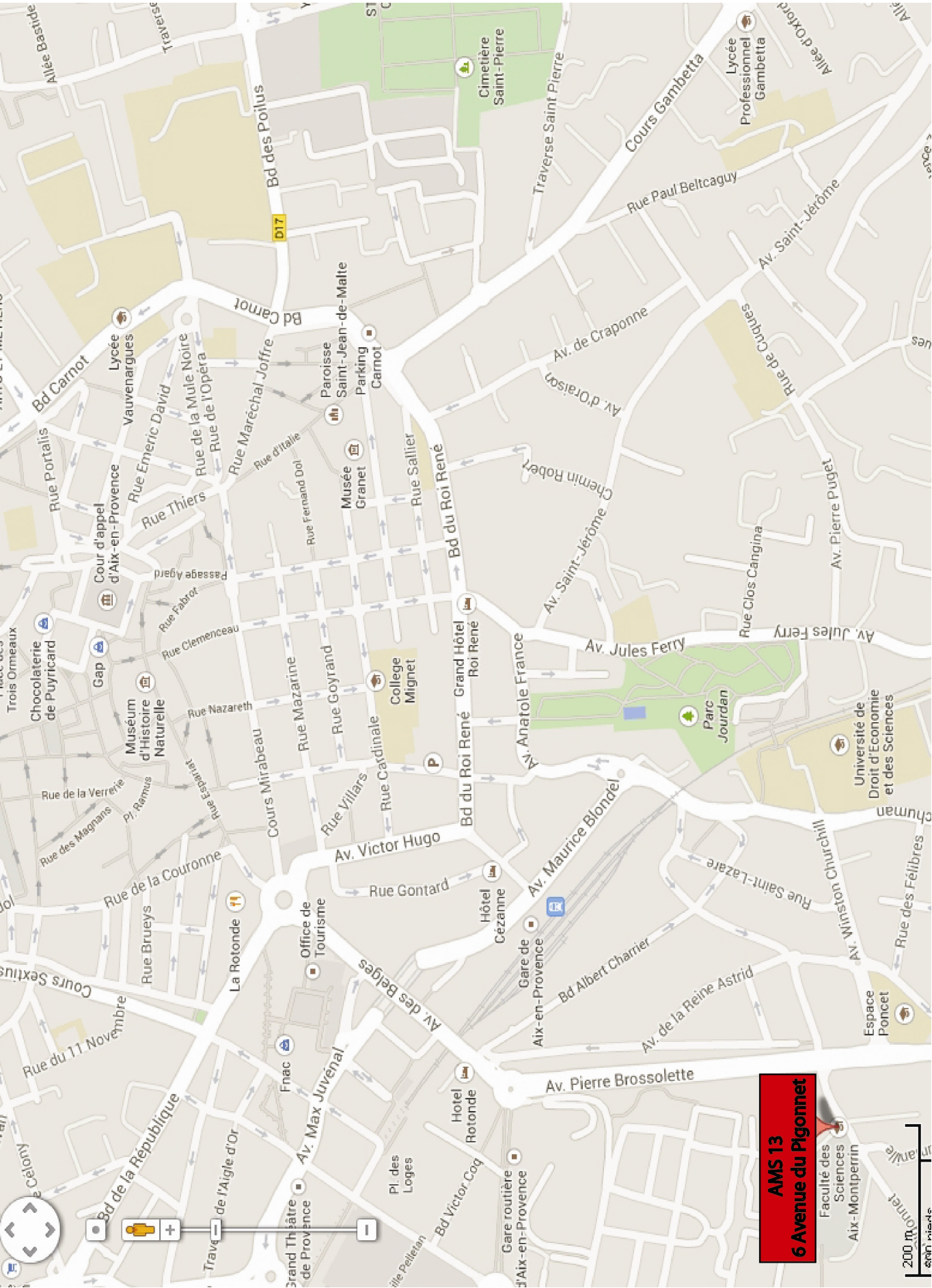
24–29 August 2014  
Aix - Marseille University - Montperrin Campus  
Aix en Provence  
France

### Programme and Abstracts Handbook



#### Conference Convenors

Régis Braucher  
Didier Bourlès



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**WELCOME to AMS-13 !**

It is a pleasure to welcome you in our lovely Provence to attend the thirteenth international conference on Accelerator Mass Spectrometry taking place in Montperrin campus, kindly made available by the Aix-Marseille University.

During these five days, you will be able to meet and exchange with your colleagues and friends coming from more than 30 countries. AMS13 will gather together more than 250 scientists working in the field of Accelerator Mass Spectrometry. We encourage our youngest colleagues to take advantage of this event to share orally with experienced "senior" scientists. 305 hundred abstracts have been submitted that have been distributed among 125 oral presentations and 180 posters. We are very grateful to our colleagues from the advisory scientific panel that firstly accepted to be part of the review process and that effectively did it. This greatly helped us.

Aside the conference, a guided tour of the historical center of Aix en Provence will end up at a welcome reception offered by the municipality in the gardens of a 18th century residence : the "Pavillon Vendôme. Wednesday afternoon, you will have the opportunity to reach the "Plateau de l'Arbois" that not only hosts the TGV station but also the "CEREGE", one of the top french laboratory dedicated to the geosciences. Between your visit to "ASTER", the 5MV French AMS national facility and the newly installed Aix-Micadas, you will enjoy a buffet offered by the "Syndicat Mixte de l'Arbois" that manages the entire site. Finally, Thursday afternoon will bring you to the "Camargue" (the Rhône delta) where you will assist to regional traditional activities such as "Ferrade", Herder's game, "Course camarguaise" and and hopefully appreciate the gala dinner animated by a Gypsy show.

Once again, we hope that you will enjoy your stay in Aquae Sextiae (Aix en Provence), the Roman thermal city that also sheltered the childhood and the maturity of Paul Cézanne.

Benvenuto en Ais de Prouvènço !

Régis Braucher and Didier Bourlès.



Paul Cézanne, Le Mont Sainte-Victoire au-dessus de la route du Tholonet, 1896-98  
(The State Hermitage Museum, Saint-Petersburg)

## 1 Scientific Advisory Panel

Anjos Roberto	Brazil
Bard Edouard	France
Bourlès Didier	France
Braucher Régis	France
Christl Marcus	Switzerland
Dewald Alfred	Germany
Fedi Mariaelena	Italy
Fink David	Australia
Heinemeier Jan	Danmark
Kieser Liam	Canada
Korschinek Gunther	Germany
Merchel Silke	Germany
Nadeau Marie Josée	Germany
Ognibene Ted	USA
Possnert Göran	Sweden
Schimmelpfennig Irene	France
Smith Andrew	Australia
Wallner Anton	Australia

## 2 Local Organising Committee

Régis Braucher  
Didier Bourlès  
Georges Aumaître  
Karim Keddadouche

with the help **S. Bertin** and **G. Dingwall** from the Aix en Provence Tourism Office

### 3 Sponsors

We thank our sponsors and exhibitors for their valuable support



## 4 General information

### Registration and Information Desk

The registration and information desk will be located in the hall of the Montperrin University from 16.00 to 18.30 on Sunday 24 August and from 8.00 to 9.00 each day thereafter as well as during coffee breaks and lunches.

### Luggage

On Friday there will be a luggage storage area available for registrants.

### Name Badges

Please wear your name badges at all times to be identify as participants. The staff of the University will have the right to prevent you to access the conference site without these badges.

### Breaks and lunches

Morning teas, lunches and afternoon teas will be served in the main hall or outside depending of the weather. Please, inform us of any special any special dietary requirements.

### Internet

Free WI-FI will be available. Personal code will be attributed to each participant. Those who have an EDUROAM email address will be able to connect the network with their own password and login.

### Presenters

A quiet room with a computer will be dedicated to test you presentation. Please report to the staff in the room in which you are presenting during a break well in advance of your presentation.

Talks will be 15 min long plus 5 minutes of questions except for sessions **2A** (Monday morning), **6B** (Tuesday morning) and **12B** (Thursday morning) for which talks will be 10 minutes plus 5 minutes of questions.

**The available softwares will be : Powerpoint Office 2010 (version 14), LibreOffice 4.1.6 and Adobe reader XI.**

Plenary sessions will held in the "Grand Amphithéâtre", low level and parallel sessions will be held in the two upper level Amphitheaters (session labeled "A" in amphitheater 1 and session labeled "B" in amphitheater 2) . Poster sessions will be organized in the public area of the campus.

**Posters will be 841 mm wide by 1188 mm high (A0 size).**

The first session posters may be installed Sunday 24 August at the Icebreaker and should be in place by Monday 25 August. This poster session is planned for Tuesday after lunch. These should be removed no later than morning tea time Wednesday 27 August. The second session posters may be installed at lunch time on Wednesday 27August and this poster session is planned for Thursday after lunch. These posters should be removed at the end of the conference on Friday 29 August. On Friday evening, remaining posters will be trashed.





### Manuscript Submission

As for the previous conferences, participants at AMS-13 have the opportunity to publish their work in a NIM-B proceedings volume. The guest editors are Régis Braucher and Didier Bourlès.

The signed contract with Elsevier stipulates the review process for this special issue is conducted in an appropriate manner and in line with normal review practices for the journal. In order to facilitate the review process, Elsevier will arrange access to the Elsevier Editorial System (EES) for the Guest Editor.

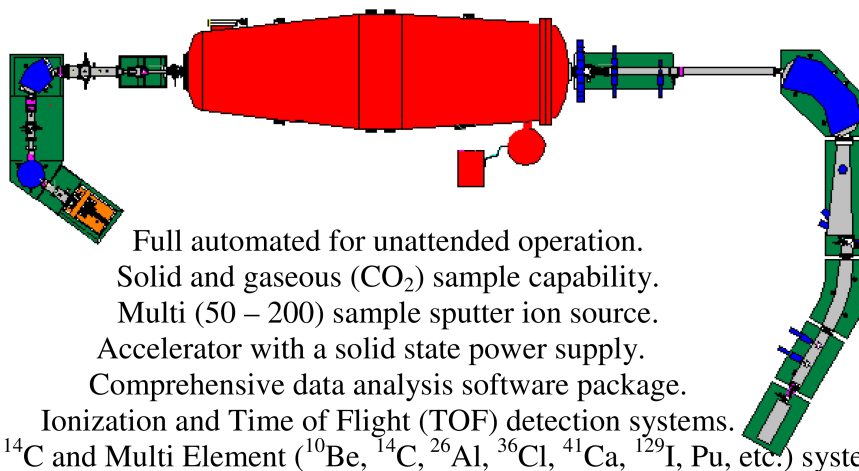
The Special Issue is expected to comprise less than 300 papers. In order to facilitate a timely publication schedule, the proposed deadline for all manuscripts to be submitted to EES is **December 1 st , 2014** and the deadline for all manuscripts to be ready for production (i.e. final decisions made on all papers and communicated to all involved parties) is June 1 st , 2015.

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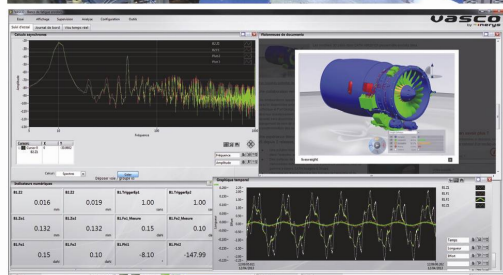
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## 5 Topic codes

Topic	Topic Code
Progress Report	PRE
New and Future Facilities	NFF
Ion Source and System Interface	ISSI
Advances in AMS Technique	AAT
AMS Carrier, Reference material and Intercomparison	CRI
Measurement Difficulties of the Normalizing Stable Isotopes	MNSI
Sample Preparation	SP
AMS and Heavy Nuclides	AHN
Forensics and Nuclear Safeguards	FNS
Combined Nuclides Applications	CNA
General AMS Applications	GAA



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## 6 Sunday 24 August 16h-18h30

Pick up of registration materials and welcome Icebreaker

## 7 Monday 25 August- Morning Sessions

### Session 1 - Grand Amphithéâtre

9h00	Opening
9h40	GAA-19 <i>Ramsey</i> AMS : Dating Methods or Correlation Tools ?
10h00	AAT-2 <i>Eliades</i> Negative ion-gas reaction studies using ion guides and AMS
10h20	SP-01 <i>Merchel</i> Improving AMS-chemistry : Two steps forward, one step back.
10h40	Break

### Session 2A - amphitheater 1 10 min. talk ; 5 min. questions

11h00	NFF-07 <i>Kieser</i> The André E. Lalonde AMS Laboratory – the new accelerator mass spectrometry facility at the University of Ottawa
11h15	NFF-09 <i>Povinec</i> A new AMS laboratory at the Comenius University in Bratislava
11h30	NFF-05 <i>Hotchkis</i> Performance of the ANSTO 1MV AMS system
11h45	NFF-14 <i>Chopra</i> A New AMS Facility at Inter University Accelerator Centre, NewDelhi, India.
12h00	NFF-15 <i>Silveira Gomes</i> LAC-UFF : The New and First $^{14}\text{C}$ -AMS Facility in Latin America
12h15	NFF-10 <i>Watrous</i> Installation of a 0.5 MV Accelerator Mass Spectrometry at Idaho National Laboratory
12h30	AAT-20 <i>Calcagnile</i> The new AMS system at CEDAD for the analysis of $^{10}\text{Be}$ , $^{26}\text{Al}$ , $^{129}\text{I}$ and actinides : set-up and performances
12h45	Lunch

### Session 2B - amphitheater 2

11h00	CNA-08 <i>Reedy</i> Factors Affecting Production Rates of Extraterrestrial Cosmogenic Nuclides
11h20	GAA-25 <i>Smith</i> Studying the constancy of galactic cosmic rays using cosmogenic radionuclides and noble gases in iron meteorites.
11h40	GAA-33 <i>Ostdiek</i> Measurement the Half Life of $^{60}\text{Fe}$ for Stellar and Early Solar System Models
12h00	GAA-09 <i>Wallner</i> Search for a live supernova signature of $^{60}\text{Fe}$ in deep-sea sediments and a new half-life measurement of $^{60}\text{Fe}$
12h20	GAA-30 <i>Ludwig</i> Search for supernova produced $^{60}\text{Fe}$ in Earth's microfossil record
12h40	Lunch

## 8 Monday 25 August- Afternoon Sessions

### Session 3A - amphitheater 1

14h00	<b>ISSI-01</b> <i>Fahrni</i> Developments and applications in $^{14}\text{C}$ gas analysis : dating a sediment core on sub-mg foraminifera samples
14h20	<b>ISSI-02</b> <i>Salazar</i> Flow separation of gas components with different axial momentum for online coupling of $\text{CO}_2$ -producing analytical instruments with Accelerator Mass Spectrometry
14h40	<b>ISSI-03</b> <i>Münsterer</i> Rapid Radiocarbon Analyses by Laser Ablation
15h00	<b>ISSI-04</b> <i>Ognibene</i> An Interface for the Direct Coupling of Small Liquid Samples to AMS
15h20	<b>GAA-26</b> <i>Hong</i> Age determination of metal types by measurement of "meok" using AMS
15h40	<b>Break</b>

### Session 3B - amphitheater 2

14h00	<b>GAA-34</b> <i>Gómez Guzmán</i> Production of $^{41}\text{CaH}_2$ samples for AMS measurements. Application to Interplanetary Dust Particles
14h20	<b>CNA-02</b> <i>Feige</i> Multiple radionuclide study of a recent supernova event in deep-sea sediments with AMS
14h40	<b>GAA-15</b> <i>Fimiani</i> Evidence for the deposition of interstellar material on the lunar surface
15h00	<b>NFF-08</b> <i>Schiffer</i> A dedicated AMS setup for $^{53}\text{Mn}/^{60}\text{Fe}$ at the Cologne FN Tandem Accelerator
15h20	<b>AAT-21</b> <i>Famulok</i> Application of AMS for the Analysis of Primordial Nuclides in High Purity Copper
15h40	<b>Break</b>



## Monday 25 August- Afternoon Sessions ; continuing

### Session 4A - amphitheater 1

<b>16h00</b>	<b>SP-11</b> <i>Lifton</i> Progress in automated extraction and purification of in situ $^{14}\text{C}$ from quartz : results from the Purdue in situ $^{14}\text{C}$ laboratory
<b>16h20</b>	<b>CRI-01</b> <i>Wacker</i> Improving the tree-based radiocarbon calibration curve
<b>16h40</b>	<b>FNS-03</b> <i>Nakamura</i> Age estimation based on an annual variation curve of bomb-produced radiocarbon concentration : growth process of an elephant tusk
<b>17h00</b>	<b>GAA-03</b> <i>Kretschmer</i> Compound specific radiocarbon analysis from indoor air samples via accelerator mass spectrometry
<b>17h30</b>	<b>Aix guided visit and Pavillon Vendôme reception</b>

### Session 4B - amphitheater 2

<b>16h00</b>	<b>CNA-09</b> <i>Dunai</i> Aeolian soil loss from agricultural areas as monitored by fallout plutonium
<b>16h20</b>	<b>CRI-03</b> <i>Rood</i> Towards high precision and low ratio $^{10}\text{Be}$ measurements with the SUERC 5MV tandem : bigger isn't always better
<b>16h40</b>	<b>GAA-01</b> <i>Welten</i> Evidence for cosmic-ray spike at 774/5 AD in annual resolution $^{10}\text{Be}$ record of West Antarctic ice core
<b>17h00</b>	<b>GAA-40</b> <i>Shiroya</i> Long-term hillslope erosion rates of Yakushima Island, southern Japan deduced from cosmogenic $^{10}\text{Be}$ in river sediments
<b>17h30</b>	<b>Aix guided visit and Pavillon Vendôme reception</b>

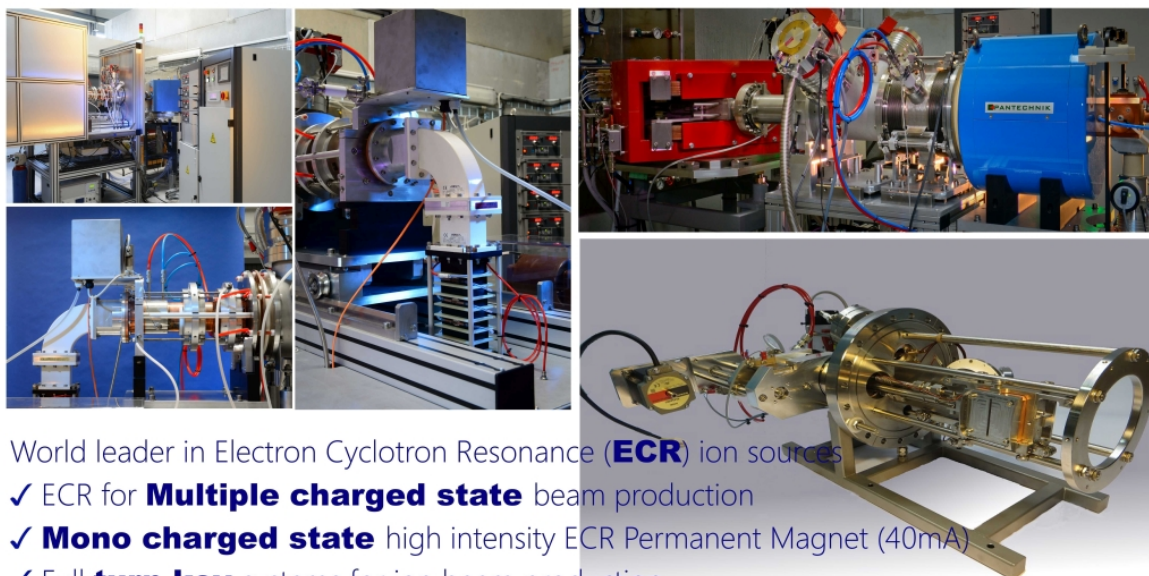
## 9 Tuesday 26 August- Morning Sessions

### Session 5 - Grand Amphithéâtre

9h00	AAT-15 <i>Synal</i> Progress in design and performance of low energy AMS systems
9h20	AAT-13 <i>Müller</i> $^{26}\text{Al}$ measurements below 500 kV
9h40	AAt-14 <i>Fu</i> Further improvement for $^{10}\text{Be}$ measurements with an upgraded compact radiocarbon facility
10h00	SP-07 <i>Fedi</i> Memory effects using an elemental analyser to combust radiocarbon samples : failure and recovery
10h20	SP-08 <i>Paul</i> Systematic investigation on contamination of background AMS samples
10h40	Break brought to you by PANTECHNIK



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## Session 6A - amphitheater 1

11h00	<b>CNA-01</b> <i>Hain</i> Identification of Actinides Emitted into the Pacific Ocean by the Fukushima Accident
11h20	<b>FNS-02</b> <i>Shibata</i> Iodine-129 and other radionuclides in the atmosphere at Tsukuba, Japan, after the Fukushima Daiichi Nuclear Power Plant accident
11h40	<b>GAA-11</b> <i>Bautista</i> Study on the iodine transportation in the fluvial system by analysis of iodine-129 concentration in water samples collected around the Fukushima Daiichi Nuclear Power Plant
12h00	<b>GAA-27</b> <i>Matsunaka</i> Depth profiles of $^{129}\text{I}$ and $^{129}\text{I} / ^{127}\text{I}$ ratio in soil at the near-field site of the Fukushima Dai-ichi Nuclear Power Plant
12h20	<b>GAA-36</b> <i>Honda</i> Speciation analysis of Iodine-127 and 129 in the soil contaminated by the Fukushima Dai-ich Nuclear Power Plant accident
12h40	<b>Lunch</b>

## Session 6B - amphitheater 2 10 min. talk ; 5 min. questions

11h00	<b>NFF-03</b> <i>Nadeau</i> Status of the "new" AMS facility in Trondheim
11h15	<b>NFF-11</b> <i>Heinemeier</i> The New HVE 1 MV Multi-Element AMS system installed at the Aarhus AMS Dating Centre
11h30	<b>NFF-01</b> <i>Zoppi</i> Performance evaluation of the new DirectAMS geochronological sample preparation laboratory
11h45	<b>NFF-02</b> <i>Chavez</i> "Laboratorio de Espectrometría de Masas con Aceleradores" (LEMA) : New AMS Facility in Mexico
12h00	<b>NFF-04</b> <i>Sasa</i> The new 6 MV multi-nuclide AMS facility at the University of Tsukuba
12h15	<b>NFF-06</b> <i>Fink</i> The new AMS Centre for Accelerator Science at ANSTO - a vision to the future
12h30	<b>SP-06</b> <i>Horiuchi</i> Development of ultrasensitive $^{10}\text{Be}$ analysis at MALT
12h45	<b>Lunch</b>

## 10 Tuesday 26 August- Afternoon Sessions

### POSTER Session 1

14h00	All AAT, AHN, CNA, CRI and FNS posters, and GAA5 to GAA 77
15h40	Break

### Session 7A - amphitheater 1

16h00	<b>AHN-06</b> <i>Jull</i> Environmental Iodine-129 studies at the University of Arizona
16h20	<b>GAA-07</b> <i>Chamizo</i> Presence of $^{236}\text{U}$ in an abyssal sediment core from the North Atlantic
16h40	<b>GAA-10</b> <i>Zhang</i> Speciation of $^{129}\text{I}$ and $^{127}\text{I}$ in Seawater from the Arctic
17h00	<b>GAA-14</b> <i>López-Gutiérrez</i> Recent evolution of $^{129}\text{I}$ levels in the Arctic and North Atlantic Oceans
17h20	<b>GAA-35</b> <i>Hou</i> Water circulation in the Norwegian Sea and the Arctic traced by AMS analysis of iodine-129
17h40	<b>SP-05</b> <i>Kusuno</i> An approach for measurement of extremely low $^{129}\text{I}$ concentration in marine fish samples

### Session 7B - amphitheater 2

16h00	<b>GAA-04</b> <i>Winkler</i> Measurement of Chlorine as trace element at the ppb-level by combining neutron activation and accelerator mass spectrometry of $^{36}\text{Cl}$
16h20	<b>GAA-32</b> <i>Miyake</i> Measurement of $^{36}\text{Cl}$ in surface soil around F1NPP accident site
16h40	<b>SP-03</b> <i>Pupier</i> Optimization of the Isotope Dilution-Accelerator Mass Spectrometry (ID-AMS) technique to analyze waters with low chlorine contents
17h00	<b>GAA-23</b> <i>Casacuberta</i> $^{236}\text{U}$ , $^{129}\text{I}$ and Pu-isotopes as oceanographic tracers in the Arctic and the Atlantic Ocean
17h20	<b>FNS-04</b> <i>Srncik</i> Bomb-produced $^{236}\text{U}$ and $^{239,240}\text{Pu}$ from a modern coral surviving the nuclear testing period at Enewetak Atoll (Marshall Islands)
17h40	<b>GAA-06</b> <i>Tims</i> $^{236}\text{U}$ and $^{239,240}\text{Pu}$ ratios from soils around an Australian nuclear weapons test site



# 11 Wednesday 27 August- Morning Sessions

## Session 8 - Grand Amphithéâtre

9h00	<b>AAT-04</b> <i>Suter</i> What can we learn from modeling the physics of AMS ?
9h20	<b>AAT-22</b> <i>Alary</i> Isobar Separator for Anions : Current Status
9h40	<b>AAT-05</b> <i>Vockenhuber</i> Isobar-separation techniques for 6 MV Tandem accelerators
10h00	<b>AAT-07</b> <i>Forstner</i> The ILIAS project for selective isobar suppression by Laser photodetachment
10h20	<b>AAT-18</b> <i>Zhao</i> Studies of the intrinsic ion transmission of RF ion guides for AMS : I
10h40	<b>Break</b>

## Session 9A - amphitheater 1

11h00	<b>AAT-08</b> <i>Martschini</i> Isobar separation of $^{93}\text{Zr}$ and $^{93}\text{Nb}$ at 24MeV with a new multi-anode ionization chamber
11h20	<b>FNS-01</b> <i>Lu</i> Development of $^{93}\text{Zr}$ - Nb separation for future AMS measurement
11h40	<b>AHN-09</b> <i>Sookdeo</i> Determining $^{210}\text{Pb}$ by accelerator mass spectrometry
12h00	<b>GAA-20</b> <i>Christl</i> The distribution of $^{236}\text{U}$ and $^{129}\text{I}$ in the North Sea in 2009
12h20	<b>PRE-09</b> <i>He</i> Measurement $^{59}\text{Ni}$ and $^{63}\text{Ni}$ by accelerator mass spectrometry at CIAE
12h40	<b>Lunch</b>

## Session 9B - amphitheater 2

11h00	<b>AAT-06</b> <i>Freeman</i> Radiocarbon positive-ion accelerator mass spectrometry
11h20	<b>AAT-17</b> <i>Seilerman</i> Status on mass spectrometric radiocarbon detection at ETHZ
11h40	<b>AAT-16</b> <i>McCartt</i> Quantification of $^{14}\text{C}$ with Cavity Ring-Down Spectroscopy
12h00	<b>GAA-02</b> <i>Soulet</i> Developments in ramped-combustion radiocarbon analysis of natural sediment : towards correcting organic carbon composition ( $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ ) for carbonate contribution
12h20	<b>SP-10</b> <i>Yang</i> Second generation laser-heated microfurnace for graphitisation of microgram samples
12h40	<b>Lunch</b>

## 12 Wednesday 27 August- Afternoon Sessions

### Session 10A - amphitheater 1

14h00	<b>PRE-04</b> <i>Fifield</i> Upgraded isotope-cycling system for the 14UD Pelletron accelerator at the Australian National University
14h20	<b>PRE-08</b> <i>Korschinek</i> AMS with heavy nuclides at the Munich Tandem accelerator
14h40	<b>SP-09</b> <i>Watrous</i> Electrodeposition as an alternate method for preparation of environmental samples for iodide for analysis by AMS
15h00	<b>AAT-09</b> <i>Fedi</i> Preliminary measurements on the new TOF system installed at the AMS beam line of INFN-LABEC
15h20	<b>AAT-19</b> <i>Stan-Sion</i> AMS method for depth profiling of trace elements concentration in materials - construction and applications
15h40	<b>GAA-22</b> <i>Lubritto</i> AMS radiocarbon dating of mortar and plaster : the case study of the Modena medieval unesco site
16h00	<b>Into bus ASTER ; Aix-Micadas Visits and Break !</b>

### Session 10B - amphitheater 2

14h00	<b>GAA-31</b> <i>Buchholz</i> How old is the human heart ?
14h20	<b>GAA-41</b> <i>Quarta</i> Bringing AMS radiocarbon into the anthropocene : potential and drawbacks in the determination of the bio-fraction in industrial emissions and in carbon-based products
14h40	<b>PRE-05</b> <i>Fueloep</i> Progress report on a novel in-situ $^{14}\text{C}$ extraction scheme at the University of Cologne
15h00	<b>SP-02</b> <i>Southon</i> A tale of tar : collagen extraction from asphalt-impregnated bones
15h20	<b>SP-04</b> <i>Gulliver</i> Extraction and analysis of sub-milligram per litre concentrations of Methane from ground-water for $^{14}\text{C}$ analysis
15h40	<b>CRI-06</b> <i>Scott</i> Continuing developments in the $^{14}\text{C}$ community inter-comparisons (SIRI)
16h00	<b>Into bus ASTER ; Aix-Micadas Visits and Break !</b>

## 13 Thursday 28 August- Morning Sessions

### Session 11 - Grand Amphithéâtre

9h00	<b>ISSI-06</b> <i>Vogel</i> Anion Formation by Neutral Resonant Ionization
9h20	<b>AAT-01</b> <i>Uribarri</i> Ion Source Development for Ultratrace Detection of Uranium and Thorium
9h40	<b>NFF-16</b> <i>Pavetich</i> From tip to toe – Improvements of the DREAMS facility for the determination of volatile and heavy radionuclides
10h00	<b>CRI-05</b> <i>Hou</i> Preparation of carrier free iodine target for speciation analysis of $^{129}\text{I}$ in environmental samples by AMS
10h20	<b>AAT-03</b> <i>Charles</i> I/Te separation in an RFQ gas cell and the potential use of $^{125}\text{I}$ as a spike for AMS analysis of $^{129}\text{I}$ at low levels
10h40	Break brought to you by NEC

# Compact AMS Systems

National Electrostatics Corp. offers a wide variety of compact, low voltage AMS systems for radio isotope ratio measurement through the actinides. NEC also provides complete AMS systems up to 25MV. All NEC systems provide high precision and low background. They can be equipped with a high throughput, multi-sample ion source or dual ion source injector for added versatility.

**Available Isotopes: C, Be, Al, Ca, I, Actinides**

Model	Isotopes	Terminal Voltage (MV)
SSAMS	C	0.25
CAMS	C	0.50
XCAMS	Be, C, Al	0.50
UAMS	Be, C, Al, Ca	1.00
IAMS	C, I	0.50
Actinide AMS	actinides	1.00

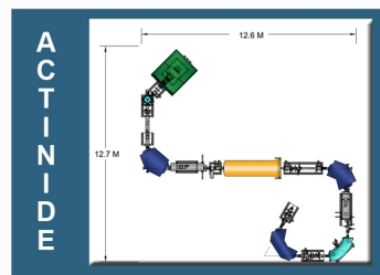
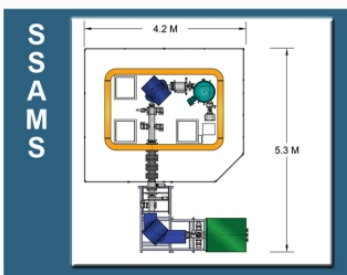
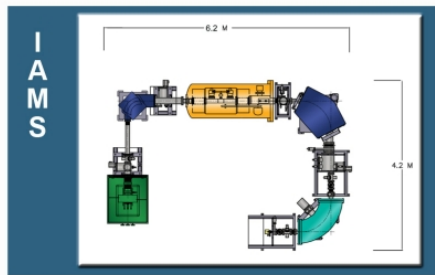
### Features:

**Better than 3 per mil precision and better than  $1 \times 10^{-15}$  background for  $^{14}\text{C}/^{12}\text{C}$**

**Gas and solid sample sources available**

**All Metal/Ceramic Acceleration tubes with no organic material in the vacuum volume**

**Automated Data Collection and Analysis**



## Session 12A - amphitheater 1

11h00	<b>GAA-29</b> <i>Sakurai</i> Induced nuclide $^{10}\text{Be}$ , $^{26}\text{Al}$ , and $^{22}\text{Na}$ in a granite core exposed by 160 GeV/c muon
11h20	<b>GAA-18</b> <i>Granger</i> Production rate of $^{10}\text{Be}$ in magnetite
11h40	<b>GAA-38</b> <i>Heikkilä</i> $^{10}\text{Be}$ in polar ice as proxy or solar activity
12h00	<b>GAA-12</b> <i>Baroni</i> Solar activity over the last millennium based on a new $^{10}\text{Be}$ record from Dome C (Antarctica)
12h20	<b>GAA-21</b> Moved to poster session 1 <i>Fujioka</i> Long-term waterfall dynamics in monsoonal Australia based on cosmogenic $^{10}\text{Be}$
12h40	<b>Lunch</b>

## Session 12B - amphitheater 2 10 min. talk ; 5 min. questions

11h00	<b>PRE-03</b> <i>Lu</i> Status of the 3MV multi-element AMS in Xi'an, China
11h15	<b>PRE-06</b> <i>Heinze</i> The first three years of CologneAMS
11h30	<b>PRE-07</b> <i>Santos</i> Status report of the 1 MV AMS facility at CNA
11h45	<b>PRE-01</b> <i>Southon</i> Equipment upgrades at the UC Irvine Keck AMS laboratory
12h00	<b>PRE-02</b> <i>Shanks</i> Performance of the rebuilt SUERC single-stage accelerator mass spectrometer
12h15	<b>MNSI-01</b> <i>Linares</i> Radiocarbon measurements at LAC-UFF : recent performance
12h30	<b>Lunch</b>



## 14 Thursday 28 August- Afternoon Session

### POSTER Session 2

14h00	GAA-78 to GAA-106, All ISSI, MNSI, NFF, PRE and SP posters
15h40	Break
16h00 - 24h 00	Into Bus, Manade Jacques Bon, Exhibition and Dinner.

**Ionplus<sup>+</sup>**  
engineering scientific instruments

In 2013, **Ionplus<sup>+</sup>** was incorporated as a spin-off company by the Laboratory of Ion Beam Physics at ETH Zurich. Based on years of experience and research, we develop, build and distribute innovative instruments for radiocarbon sample preparation and measurements. Our products are designed for fast and efficient processing of samples and cover virtually the entire range of dedicated  $^{14}\text{C}$  laboratory equipment. **Ionplus<sup>+</sup>** offers fully automated graphitization systems (AGE), gas interface systems (GIS), automated carbonate handling systems, pneumatic sample presses and vacuum lines for sealing tubes and a range of accessories for all products. Our

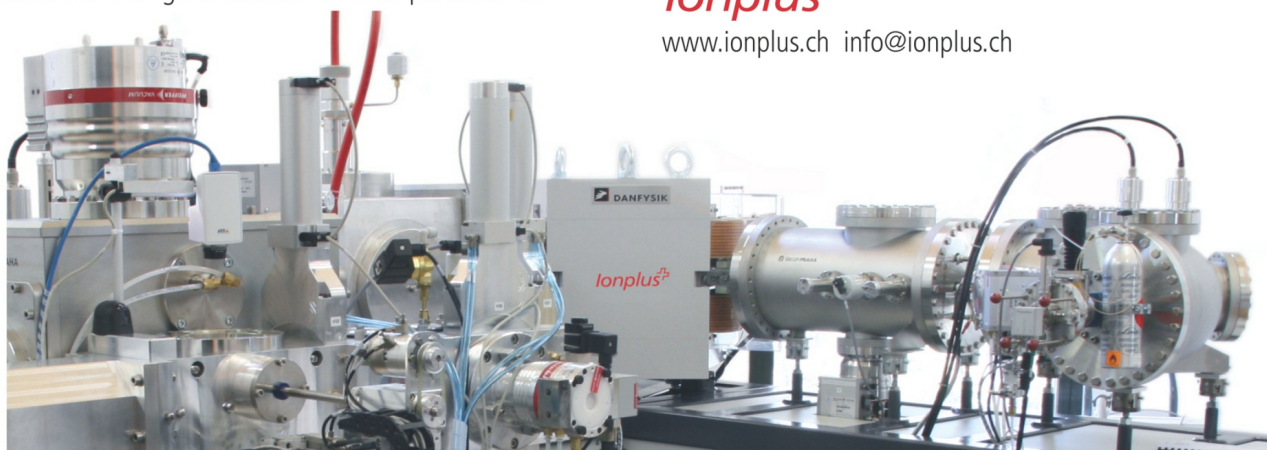
goal for these products is a high degree of automation and versatility, as well as a compact and user-friendly design achieved through excellent engineering.

In 2016, **Ionplus<sup>+</sup>** will take over the production of the Mini Carbon Dating System (MICADAS) from ETH Zurich, where 8 instruments have been built and delivered to customers in Europe and the US so far. Thus, the most compact  $^{14}\text{C}$ -AMS system commercially available will complete our product range.

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## 15 Friday 27 August- Morning Sessions

### Session 13 - Grand Amphithéâtre

9h00	<b>AHN-02</b> <i>Galindo-Uribarri</i> Accelerator Mass Spectrometry applied to measure trace levels of actinides in underground detector experiments
9h20	<b>AHN-05</b> <i>Wallner</i> A novel method for studying neutron-induced reactions on actinides
9h40	<b>AHN-10</b> <i>Dai</i> Improved target preparation methods for actinides by AMS
10h00	<b>AHN-11</b> <i>Bauder</i> The Use of Laser Ablation in an Electron Cyclotron Resonance Ion Source for Actinide Detection by AMS
10h20	<b>AHN-03</b> <i>Buompane</i> Background reduction in $^{236/238}\text{U}$ measurements
10h40	<b>Break</b>

### Session 14A - amphitheater 1

11h00	<b>CRI-04</b> <i>Cornett</i> Actinide Measurements by AMS and AS using Fluoride Matrices
11h20	<b>AHN-07</b> <i>Pardo</i> Ultra-High Sensitivity Techniques for the Determination of $^3\text{He}/^4\text{He}$ Abundances in Helium by Accelerator Mass Spectrometry
11h40	<b>AHN-01</b> <i>Steier</i> The AMS isotope Uranium-236 at VERA
12h00	<b>AHN-04</b> <i>De Cesare</i> A new fast-cycling system for AMS at ANU
12h20	<b>AHN-08</b> <i>Sakaguchi</i> Reconstruction of anthropogenic $^{236}\text{U}$ input to the Japan Sea
12h40	<b>Lunch</b>

### Session 14B - amphitheater 2

11h00	<b>CNA-03</b> <i>Fink</i> Antarctica at the global "Last Glacial Maximum" - what can we learn from cosmogenic $^{10}\text{Be}$ and $^{26}\text{Al}$ exposure ages?
11h20	<b>CNA-05</b> <i>Fujioka</i> An inherited cosmogenic burial signal from surface dune sands in the Simpson Desert dunefield, central Australia
11h40	<b>CNA-06</b> <i>Zhou</i> New $^{10}\text{Be}$ evidence for Brunhes/Matuyama magnetic polarity reversal in Chinese Loess Plateau
12h00	<b>CNA-07</b> <i>Wirsig</i> Combined cosmogenic $^{10}\text{Be}$ and $^{36}\text{Cl}$ nuclide concentrations constrain subglacial erosion rates
12h20	<b>GAA-24</b> <i>Goerhing</i> A Bayesian approach to estimating in situ cosmogenic nuclide production rates that explicitly considers erosion
12h40	<b>Lunch</b>

## 16 Friday 29 August- Afternoon Session

### Session 15 - Grand Amphithéâtre

14h00	<b>CRI-02</b> <i>Nishiizumi</i> Preparation of New Sets of $^{10}\text{Be}$ and $^{26}\text{Al}$ AMS Standards
14h20	<b>GAA-37</b> <i>Shen</i> Study on $^{41}\text{Ca}$ -AMS technology for early diagnosis of cancer bone metastasis
14h40	<b>GAA-28</b> <i>MacDonald</i> Development of a Cs Isotope Measurement Technique for AMS
15h00	<b>Summary</b> <i>Madame ? Monsieur ? Madame et Monsieur ?</i>
15h40	<b>Break</b>

### Session 16 - Grand Amphithéâtre

16h00	AMS14 bids
16h30	Instructions for NIM B proceedings
17h00	Closing



## 17 Abstracts, in order of presentation

### 17.1 Monday 25 August - Morning

Topic : GAA 19

Session 1

#### AMS : Dating Methods or Correlation Tools ?

Bronk Ramsey Christopher,<sup>1</sup>

[1]Oxford Radiocarbon Accelerator Unit, University of Oxford (United Kingdom)

One of the main achievements of Accelerator Mass Spectrometry (AMS) has been in the area of dating : the huge explosion in the application of  $^{14}\text{C}$  dating, and in the development of new techniques such as cosmogenic isotope exposure dating. These methods are used for the generation of chronologies that are then used in other disciplines. However, generating reliable chronologies requires a holistic understanding of processes involving the radio-isotopes. This is clear in the case of cosmogenic isotope exposure dating where AMS provides data that must be used to generate a self-consistent model of erosion and geophysical change, rather than 'dates' as such. Because of the need for calibration,  $^{14}\text{C}$  is strictly a correlation rather than a dating tool. However, calibration curves are not enough to provide the full environmental context for a number of reasons. Generation of such curves needs a better knowledge of the geophysics than we have (particularly for the oceans pre-Holocene) ;  $^{14}\text{C}$  can help develop this knowledge and calibration curves should only be one output of a major exercise to model the carbon cycle over the last 50ka. Also  $^{14}\text{C}$  is rarely used in isolation, and chronologies rely on a range of 'dating' and correlation methods ; AMS has a role here too : for example by linking the chronologies of ice cores with those from  $^{14}\text{C}$ , through  $^{10}\text{Be}$ . Finally, many biological systems derive carbon from different reservoirs requiring the use of  $^{14}\text{C}$ , stable isotope and other information to resolve chronological and biological problems together. This paper will look at ways in which the research community involved with AMS might engage with the major research task of correlating processes (geophysical, biological and anthropogenic) over the Quaternary.

Topic : AAT 02

Session 1

**Negative ion-gas reaction studies using ion guides and AMS.****Eliades John,<sup>1</sup> Zhao Xiaolei,<sup>2</sup> Litherland Albert,<sup>3</sup> Kieser William.<sup>1</sup>**

[1]Korea Institute of Science and Technology,(South Korea)

[2]Physics and A. E. Lalonde Lab, Univ. of Ottawa, (Canada)

[3]Department of Physics, University of Toronto, (Canada)

Fundamental obstacles in many gas-phase negative ion reaction studies are ion production and especially unambiguous identification of the reaction products. The  $\text{Cs}^+$  sputter ion sources typical to accelerator mass spectrometry (AMS) systems readily produce usable currents of a wide variety of negative ions. Tandem accelerator AMS systems can be used for unambiguous ion identification at MeV energies, especially for the lighter elements, and AMS is a very sensitive form of mass spectrometry with the largest dynamic range of at least  $10^{15}$ . A prototype radio frequency quadrupole (RFQ) ion guide was installed in the low energy beam line, after a magnetic analyzer, on the IsoTrace Laboratory (Toronto, Canada) AMS system for studies of low kinetic energy ( $< 15$  eV) isobar suppression through ion-gas interactions. As an offshoot to that work, it was found that this RFQ-AMS system also has great potential for gas-phase negative ion reaction studies. Preliminary measurements taken as part of doctoral work are presented to demonstrate the technique. The reaction products  $\text{NO}_2^-$ ,  $\text{SO}^-$ ,  $\text{SO}_2^-$ ,  $\text{NS}^-$ , and  $\text{NSO}_2^-$  were identified when  $\text{S}^-$  was injected into the RFQ with  $\text{NO}_2$  gas after deceleration to about  $> 6$  eV kinetic energy, with greater than 6 orders of magnitude S- attenuation observed. After deceleration to  $> 5$  eV, the superhalogen ion  $\text{SrF}_3^-$  was found to be largely un-reactive with  $\text{NO}_2$  while both  $\text{ZrF}_3^-$  and  $\text{YF}_3^-$  were found to be highly reactive through  $\text{NO}_2^-$  production and oxygen capture. The novel reaction product  $(\text{YF}_3^-)\text{NO}_2$  was also observed. On the other hand,  $\text{S}^-$  was found to be largely un-reactive with  $\text{N}_2\text{O}$  despite the existence of a highly exothermic  $\text{SO}^-$  reaction channel. Considerations for future work using this type of system are also discussed.

**Improving AMS-chemistry : Two steps forward, one step back.**

Merchel Silke,<sup>1</sup> Bourlès Didier,<sup>2</sup> Feige Jenny,<sup>3</sup> Ludwig Peter,<sup>4</sup> Pavetich Stefan,<sup>1</sup> Ritter Aline,<sup>1</sup>  
Rodrigues Dario,<sup>5,6</sup> Rugel Georg,<sup>1</sup> Smith Thomas,<sup>7</sup> Ziegenrucker René.<sup>1</sup>

[1]Helmholtz-Zentrum Dresden-Rossendorf, (Germany)

[2]UM 34 Aix-Marseille Univer., CNRS - IRD , CEREGE, (France)

[3]Vienna Environmental Research Accelerator, (Austria)

[4]Technical University Munich, (Germany)

[5]Consejo Nacional de Investigaciones Científicas y Técnicas, (Argentina)

[6]Comisión Nacional de Energía Atómica, (Argentina)

[7]University of Bern, Space Research and Planetary Sciences, (Switzerland)

The DREAMS (DREsden AMS) facility consists of a sophisticated 6 MV accelerator system [1], but also provides two chemistry laboratories for external users. One lab is used for preparation of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{41}\text{Ca}$ ,  $^{53}\text{Mn}$  and  $^{60}\text{Fe}$  targets. The other one is dedicated to halide targets ( $^{36}\text{Cl}$ ,  $^{129}\text{I}$ ), thus, any use of Cl- or S-compounds such as HCl or  $\text{H}_2\text{SO}_4$  is strictly prohibited. Separation protocols are applied to calcite- and quartz-rich samples for in-situ projects [2-4]. Atmospheric  $^{10}\text{Be}$  has been leached from marine sediments and Mn-nodules for dating purposes [5] and chemistry refined for bigger samples and heavier nuclides ( $^{26}\text{Al}$ ,  $^{53}\text{Mn}$ ,  $^{60}\text{Fe}$ ) [6]. After adapting standard protocols [7] introducing simple Mn-separation by delayed hydroxide separation, higher isobar concentrations ( $^{53}\text{Cr}$ ) have been found asking for further cleaning by ion exchange. With intent to speed-up and simplify the separation procedures for ice and meteorite samples [8], difficulties have been arisen e.g. carryover of  $\text{Ag}^+$  ions into  $\text{MnO}_2$ , and shortly after overcome. Another chemistry challenge probably mastered is the dissolution of meteoritic troilite ( $\text{FeS}$ ) without losing  $\text{natCl}^-$  carrier before equilibrium with  $^{36}\text{Cl}$ , plus suppression of massive isobar amounts. Remaining issues also influencing the quality of AMS-data, such as incorrectly measured stable isotope concentrations ( $^9\text{Be}$ ,  $^{27}\text{Al}$ ), are usually underestimated and harder to tackle.

[1] Akhmadaliev et al. NIMB 294 (2013) 5.

[2] Merché et al. Quat. Geo. 22 (2014) 33.

[3] Zech et al. Paleo<sup>3</sup> 369 (2013) 253.

[4] Yildirim et al. Tectonics 32 (2013) 1107.

[5] Feige et al., Ludwig et al. & Rodrigues et al. AMS-13.

[6] Feige et al. EPJ Web Conf. 63 (2013) 03003.

[7] Merché & Herpers, RCA 84 (1999) 215.

[8] Smith et al. AMS-13.

Topic : NFF 07

Session 2A

**The André E. Lalonde AMS Laboratory – the new accelerator mass spectrometry facility at the University of Ottawa.**

Kieser William,<sup>1</sup> Clark Ian,<sup>2</sup> Cornett Jack,<sup>2</sup> Litherland Albert,<sup>3</sup> Zhao Xiaolei,<sup>1</sup> Klein Matthias,<sup>4</sup> Mous Dirk,<sup>4</sup> Alary Jean-François.<sup>5</sup>

[1]Dept. of Physics and A. E. Lalonde Lab, Univ. of Ottawa, (Canada)

[2]Dept. of Earth Sciences and A. E. Lalonde Lab, Univ. of Ottawa, (Canada)

[3]Department of Physics, University of Toronto, (Canada)

[4]High Voltage Engineering B.V., (Netherlands)

[5]Isobarex Corp, (Canada)

The University of Ottawa, Canada, has installed a multi-element, 3 MV tandem AMS system as the cornerstone of the André E. Lalonde Accelerator Mass Spectrometry Laboratory, located in their new Advanced Research Complex (ARC). Manufactured by High Voltage Engineering Europa BV, the Netherlands, it is equipped with a 200 sample ion source, a high resolution, 120° injection magnet (mass-energy product 12 MeV-AMU), a 90° high energy analysis magnet (mass-energy product 350 MeV-AMU), a 65°, 1.7m radius electric analyzer and a 2 channel gas ionization detector. It is designed to analyze isotopes ranging from tritium to the actinides and to include the use of fluoride target materials. A second injection line, consisting of selected components from the IsoTrace Laboratory, University of Toronto is being added and will contain a pre-commercial version of the Isobar Separator for Anions, manufactured by Isobarex Corp. Bolton, Ontario, Canada. This instrument uses selective ion-gas reactions in a radio-frequency quadrupole cell to attenuate both atomic and molecular isobars. Four new preparation laboratories are located in the ARC building for radiocarbon, radio-halide, tritium and actinide samples. Radiocarbon labs at Université Laval, Québec and Université de Québec at Montréal and a cosmogenic radioisotope lab at Dalhousie University, Halifax, Nova Scotia will also provide samples. This presentation will focus on the details of the new AMS equipment.



Topic : NFF 09

Session 2A

**A new AMS laboratory at the Comenius University in Bratislava.**

Povinec Pavel,<sup>1</sup> Masarik Jozef,<sup>1</sup> Holý Karol,<sup>1</sup> Ješkovský Miroslav,<sup>1</sup> Breier Robert,<sup>1</sup> Šivo Alexander,<sup>1</sup>  
Staníček Jaroslav,<sup>1</sup> Kaizer Jakub,<sup>1</sup> Pánik Ján,<sup>1</sup> Zeman Jakub.<sup>1</sup>

[1]Comenius University, (Slovakia)

A Centre for Nuclear and Accelerator Technologies (CENTA) has been established at the Comenius University in Bratislava comprising of a tandem laboratory designed for the Accelerator Mass Spectrometry (AMS) and Ion Beam Analysis (IBA) studies. The laboratory is equipped with two ion sources - Alphasource (RF source for H and He ions) and MC-SNICS (target wheel with 40 positions for solid targets), 3 MV Pelletron with nitrogen stripping column, and analyzers of accelerated ions (all equipment of National Electrostatics Corp. USA). The first studies included optimization of the ion sources for different targets, optimization of nitrogen pressure in the stripping column for the highest transition of ions with used charge, and optimization of parameters of the high energy ion analyzer. A cryogenic-vacuum line has been constructed and production of graphite targets using external and internal hydrogen supply for radiocarbon measurements has been studied. The scientific program of the CENTA will be devoted mainly to nuclear, environmental, life and material sciences.

Topic : NFF 05

Session 2A

**Performance of the ANSTO 1MV AMS system.**

Hotchkis Michael,<sup>1</sup> Wilcken Klaus,<sup>1</sup> Child David,<sup>1</sup> Fink David,<sup>1</sup> Levchenko Vladimir,<sup>1</sup> Smith Andrew,<sup>1</sup> Hauser Thilo,<sup>2</sup> Kitchen Richard.<sup>2</sup>

[1]Australian Nuclear Science and Technology Organisation, (Australia)

[2]National Electrostatics Corporation, (United States)

In October 2013 a new 1MV AMS system was installed at ANSTO, with commissioning continuing in the early part of 2014. The system has been designed to cover the full mass range, with terminal voltage and stripper gas selectable to provide optimum beam transmission for a wide range of species. Performance data for radiocarbon, actinides and other species ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{129}\text{I}$ ) will be reported.

Topic : NFF 14

Session 2A

**A New AMS Facility at Inter University Accelerator Centre, NewDelhi, India.****Chopra Sundeep,<sup>1</sup> Kanjilal Dinakar,<sup>1</sup> Kumar Pankaj,<sup>1</sup> Ojha Sunil,<sup>1</sup> Joshi Rajan,<sup>1</sup> Gargari Satinath.<sup>1</sup>**<sup>[1]</sup>Inter-University Accelerator Centre, New Delhi, (India)

Inter University Accelerator Centre (IUAC), a national facility of government of India, having 15 UD Pelletron Accelerator facility was setup for multidisciplinary Ion Accelerator based research programs. A new Accelerator Mass Spectrometry (AMS) facility has been developed after incorporating many changes in the existing 15UD Pelletron Accelerator. The major changes for modifications include installation of a bi-directional 40 cathodes MC-SNICS ion source, Recirculating Turbo-molecular pumps based gas stripper system, an offset Faraday cup after analyzer magnet, a Wien filter, a quadrupole system, double slits, gas cell and a multi-anode gas detector. The simultaneous injection of  $^9\text{Be}^{17}\text{O}$  and  $^{10}\text{Be}^{16}\text{O}$  is being done for detecting  $^{10}\text{Be}^{3+}$  in the gas ionization detector and  $^{17}\text{O}^{+5}$  in the offset Faraday cup after analyzing magnet. A maximum transmission of 19%, for 9 Be +3 charge state at a terminal potential of 10 MV, has been achieved with foil and gas stripper combination for BeO beam. A clean chemistry laboratory for  $^{10}\text{Be}$  and  $^{26}\text{Al}$  with all the modern facilities has also been developed for the chemical processing of samples. In addition a new dedicated  $^{14}\text{C}$  AMS facility is also being setup having an automated 500 kV tandem ion accelerator with two Cs sputter ion sources capable of performing measurements of samples of few mg size with a good precision. A sample preparation laboratory with automatic graphitization equipment has also been setup for the preparation of carbon samples. The new AMS facility will be completely operational by the end of year 2014.

Topic : NFF 15

Session 2A

**LAC-UFF : The New and First  $^{14}\text{C}$ -AMS Facility in Latin America.**

Silveira Gomes Paulo Roberto,<sup>1</sup> Macario K.d.<sup>1</sup> Linares R.<sup>1</sup> Carvalho C.<sup>1</sup> Santos H.c.<sup>1</sup> Castro M.d.<sup>1</sup>  
Oliveira F.m.<sup>1</sup> Mendes D.<sup>1</sup> Anjos R.m.<sup>1</sup>

[1]Instituto de Fisica, Universidade Federal Fluminense, (Brazil)

The Intitue of Physics(IF-UFF) has been developing, for more than one decade, collaborative projectsith ANU, PRIME Lab and UCI- Irvine AMS radiocarbon laboratories. A few years ago, the first Brazilian radiocarbon sample preparation laboratory for AMS technique was installed at the IF-UFF[1]. In March 2012 it started the operation at IF-UFF of a 250 kV Single Stage Accelerator Mass Spectrometry (SSAMS) system produced by NEC which, together with the preparation laboratory, put our group in an unique scenario within the Latin American radiocarbon community. Since then the number of requests for radiocarbon measurements at LAC-UFF is increasing. We deal with soil, sediments, wood, charcoal and peats from Brazilian and Latin American groups from several areas like Geosciences, Oceanography and Archaeology South. At LAC-UFF, OX-II is used as standard modern for normalization and the IAEA reference materials C2, C5 and C6 are prepared as quality control. Calcite and reactor graphite are used as standard backgrounds for inorganic and organic samples. Typical background is 0.8pMC for reactor graphite. We have already performed an inter-comparison exercise using shells, soils, vegetable fragments, charcoal and peats measured both with AMS at University of Georgia (UGAMS), and with liquid scintillation technique at CENA (Sao Paulo, Brazil) [2]. Regarding the SSAMS machine, typical currents are  $50\text{ }\mu\text{A}$   $^{12}\text{C}$ -1 measured at the low energy Faraday cup. The isotopic fractionation is corrected by measuring the  $\delta^{13}\text{C}$  on-line in the accelerator. Average machine background is 0.15 pMC and average precision is 0.8%. The transmission in the accelerator is of the order of 32 %. We have been working to extend the sample background and to increase our precision.

Topic : NFF 10

Session 2A

**Installation of a 0.5 MV Accelerator Mass Spectrometry at Idaho National Laboratory.**

Watrous Matthew,<sup>1</sup> Adamic Mary,<sup>1</sup> Delmore James,<sup>1</sup> Hague Robert,<sup>1</sup> Jenson Douglas,<sup>1</sup> Olson John,<sup>1</sup>  
Vockenhuber Christof.<sup>2</sup>

[1]Idaho National Laboratory, (United States)

[2]ETH - Zürich, (Switzerland)

A new AMS laboratory is being established with initial installation scheduled for the second half of 2014. The AMS is a National Electrostatics Corporation 0.5 MV instrument configured to analyze the iodine +3 ion. The initial application will be to replace TIMS for monitoring  $^{129}\text{I}$  at the Idaho National Laboratory (INL) that may have been released during legacy operations, such as nuclear fuel reprocessing. INL has been conducting iodine analysis via thermal ionization mass spectrometry (TIMS) beginning with the establishment of the method by James Delmore in 1982. The technique makes use of a triple filament ion source with a lanthanum hexaboride ionizing filament. With this ion source installed in a triple sector mass spectrometer the INL routinely measures the NIST SRM 3230 and NIST SRM 3231 ( $^{129}\text{I}/^{127}\text{I}$  ratios of  $4.920 \times 10^{-10}$  and  $0.982 \times 10^{-8}$ ) standards. Accelerator mass spectrometry provides an opportunity to extend this capability. There are two reasons for this transition; the sputter ion source is far less sensitive to sample purity than TIMS allowing for simplified sample chemistry and wider isotopic ratios can be measured, facilitating more accurate mapping of fission product distributions. What is now the INL began operating Nuclear Reactors in 1951 and processing irradiated nuclear fuel in 1953. Over the years INL has been home to 52 operating nuclear reactors. Testing situations have included allowing reactor cores to undergo excess heat excursions. Many different types of used fuel elements were processed between 1953 and 1989 when reprocessing ceased. These combined activities have resulted in the potential for various fission products such as  $^{129}\text{I}$  to be in the environment at very low levels.

Topic : AAT 20

Session 2A

**The new AMS system at CEDAD for the analysis of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and actinides :  
set-up and performances.**

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The Centre for Dating and Diagnostics (CEDAD) at the University of Salento was established in 2001 and became fully operational for routine  $^{14}\text{C}$  radiocarbon dating in 2003. The facility has been continuously upgraded over the years with the installation of different beamlines for high energy ion implantation, IBA analyses both in vacuum and in air and nuclear microprobe. In 2011 a second AMS beamline was installed consisting of a dedicated high energy mass spectrometer designed in collaboration with the ion beam physics group at ETH, Zurich for the AMS analysis of rare nuclides such as  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and actinides. First tests on  $^{10}\text{Be}$  allowed to optimize operating parameters resulting in the proper separation of  $^{10}\text{Be}$  from the interfering isobar  $^{10}\text{B}$ . In this paper we present the further tests and optimization which resulted in the enhancement of the overall transmission efficiency, the reduction of the background (in the  $10^{-15}$  range) and in the possibility to obtain precision levels in routine  $^{10}\text{Be}/^9\text{Be}$  measurements of the order of 0.5 %.

Furthermore the first results obtained for the analysis of  $^{26}\text{Al}$  and  $^{129}\text{I}$  are also presented.

## Factors Affecting Production Rates of Extraterrestrial Cosmogenic Nuclides

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Early in the history of cosmogenic nuclides, all production rates for a nuclide in meteorites were assumed to be constant for a given target composition. We now know that rates depend on many factors, especially the pre-atmospheric object's size, the location of the sample in that object (such as near surface or deep inside), and the object's bulk composition. Work has been done and continues to be done on better understanding those and other factors. The bulk composition affects rates, especially in objects with very low and very high iron contents. Extraterrestrial materials with high iron contents usually have higher rates for making nuclides made by spallation reactions. High iron reduces the rates for neutron-capture reactions by other elements. In small objects and near the surface of objects, the cascade of secondary neutrons is developing as primary particles are just starting to be removed. Deep in large objects that secondary cascade is fully developed and the fluxes of primary particles are low. Recent work shows that even the shape of an object in space has a small but measureable effect. With the use of modern Monte Carlo codes for the production and transport of particles, the nature of these effects have been and are being studied. Work needs to be done to improve the results of these calculations, especially the cross sections for making spallogenic nuclides.



Topic : GAA 25

Session 2B

## Studying the constancy of galactic cosmic rays using cosmogenic radionuclides and noble gases in iron meteorites.

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During their orbit in space, extraterrestrial bodies are exposed to cosmic rays. The interaction between these energetic particles and the meteoroides produce both stable and radioactive cosmogenic nuclides that can be used to study, e.g. the size of the meteorite before ablation in the Earth's atmosphere and the time the object traveled before falling on Earth (exposure age). For a proper interpretation of such data, especially the ages, the temporal constancy of the cosmic ray intensity has to be proven. Doing so and being interested in timescales in the range of a few hundred million years, we have to rely on iron meteorites because their exposure ages range from a few million to a few billion years. In this study, we systematically investigate the exposure ages of iron meteorites and search for periodic structures in the age distribution. So far, we have studied 28 iron meteorites for  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{36}\text{Cl}$  at the DREsden Accelerator Mass Spectrometry (DREAMS) facility [1] and for the noble gas isotopes of He, Ne, and Ar at the University of Bern. The first  $^{53}\text{Mn}$  and  $^{60}\text{Fe}$  measurements have already been performed at the Australian National University (ANU) and at the TUM (Munich). Finally,  $^{41}\text{Ca}$  measurements at DREAMS to identify long terrestrial residence times influencing the radionuclide concentrations are foreseen for the very near future. The measurements of additional iron meteorites, which will help improving the statistics of the age distribution as well as extending the list of radionuclides and also extending the study to mineral separates from iron meteorites, are currently ongoing.

[1] Akhmadaliev, S. et al. (2013) NIMB 294, 5.

Topic : GAA 33

Session 2B

**Measurement the Half Life of  $^{60}\text{Fe}$  for Stellar and Early Solar System Models.**

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Radioisotopes, produced in stars and ejected through core collapse supernovae (SNe), are important for constraining stellar and early Solar System (ESS) models. The presence of these isotopes, specifically  $^{60}\text{Fe}$ , can identify progenitors of SN types, give evidence for nearby SNe, and can be a chronometer for ESS events. The  $^{60}\text{Fe}$  half-life, which has been in dispute in recent years, can have an impact on calculations for the timing for ESS events, the distance to nearby SN, and the brightness of individual, non-steady state  $^{60}\text{Fe}$  gamma ray sources in the Galaxy. To measure such a long half life, one needs to simultaneously determine the number of atoms in and the activity of an  $^{60}\text{Fe}$  sample. We have undertaken a half-life measurement at the University of Notre Dame and have successfully measured the  $^{60}\text{Fe}$  concentration of our samples using Accelerator Mass Spectrometry (AMS). We will couple this result with an ongoing activity measurement using isomeric decay in  $^{60}\text{Co}$  rather than the traditional  $^{60}\text{Co}$  grow-in decay. I will present our AMS data and the most recent results of the activity measurement.

Topic : GAA 09

Session 2B

## Search for a live supernova signature of $^{60}\text{Fe}$ in deep-sea sediments and a new half-life measurement of $^{60}\text{Fe}$

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 Rugel Georg,<sup>3</sup> Schumann Dorothea,<sup>4</sup> Tims Steve,<sup>1</sup> Winkler Stephan,<sup>2</sup> Sterba Johannes,<sup>5</sup> Bichler  
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$^{60}\text{Fe}$  (2.6 My) is one of the most versatile nuclides in astrophysics. Live  $^{60}\text{Fe}$  was identified in the Galaxy. Its stellar production requires neutron densities available only in explosive Supernovae (SNe) or SuperAGB stars.  $^{60}\text{Fe}$  is also found in meteorites and indirectly via  $^{60}\text{Ni}$  anomalies. Fresh nucleosynthetic products may enter the Solar System (SS) trapped in cosmic dust. Hence, nearby SNe might deposit traces on Earth and since it has negligible terrestrial production,  $^{60}\text{Fe}$  is an ideal candidate to search for recent SNe. However, detection requires sensitivities of  $^{60}\text{Fe}/\text{Fe} \sim 10^{-15}$  and  $^{60}\text{Fe}$ -AMS faces interference from stable  $^{60}\text{Ni}$ . So far only TU Munich, combining a MP tandem with a gas-filled magnet, measures  $^{60}\text{Fe}$  routinely as low as  $\sim 10^{-16}$  [1]. This group discovered live  $^{60}\text{Fe}$  in a deep-sea crust indicating that SNe-isotopes found their way to Earth 2-3 My ago. Work is ongoing at TUM to validate this finding in other archives [2-3]. Further, Rugel et al. [4] measured a half-life substantially longer than previously recommended. We have started a similar program at the ANU using the 14UD accelerator and a split-pole magnetic spectrograph converted into a gas-filled magnet. A substantial beamtime devoted to  $^{60}\text{Fe}$  has resulted in an exceptional sensitivity below  $10^{-16}$ . We have searched for a SN-signal in 3 deep-sea sediment cores (Indian Ocean) [5]. We will present exciting new data for  $^{60}\text{Fe}$  with high time resolution and will relate it to potential exposure of the SS to recent SNe. In addition, we re-measured in an independent approach the  $^{60}\text{Fe}$  half-life via AMS measurements of the  $^{60}\text{Fe}/^{55}\text{Fe}$  ratio.

[1] Knie et al. PRL93 (2004)

[2] Ludwig et al. this conf.

[3] Fimiani et al. LPSC 1659 (2012)

[4] Rugel et al. PRL103 (2009)

[5] Feige et al. this conf.

**Search for supernova produced  $^{60}\text{Fe}$  in Earth's microfossil record**

**Ludwig Peter,<sup>1</sup> Bishop Shawn,<sup>1</sup> Egli Ramon,<sup>2</sup> Faestermann Thomas,<sup>1</sup> Famulok Nicolai,<sup>1</sup> Fimiani Leticia,<sup>1</sup> Gómez Guzmán José Manuel,<sup>1</sup> Hain Karin,<sup>1</sup> Korschinek Gunther,<sup>1</sup> Hanzlik Marianne,<sup>1</sup> Merchel Silke,<sup>3</sup> Rugel Georg.<sup>3</sup>**

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Nucleosynthesis in massive stars can produce copious amounts of the radioisotope  $^{60}\text{Fe}$  ( $T(1/2)=2.62\text{ Ma}$ ). When those stars end their lives in a supernova, material enriched with nucleosynthesis products can be ejected into the interstellar medium. If such supernova debris is picked up by Earth, it can be incorporated into terrestrial reservoirs. After the discovery of live  $^{60}\text{Fe}$  atoms in 2-3 Myr old layers of a Pacific Ocean ferromanganese crust (K. Knie et al. Phys. Rev. Lett. 93(2004) 171103), a confirmation of this signal, as well as a mapping of the signal with high time-resolution is desirable. Another reservoir in which the  $^{60}\text{Fe}$  signature should have been incorporated in are the fossils of magnetotactic bacteria in ocean sediment. These bacteria form chains of small magnetite crystals for magnetotaxis. After cell death and sedimentation, these magnetic chains can be preserved even over geologically significant timescales. In order to extract iron from secondary,  $^{60}\text{Fe}$  bearing minerals only, a carefully tuned chemical leaching technique was employed. A novel technique to characterize this procedure using magnetic measurements was also developed applied to quantify secondary magnetite in our samples. As sample materials, two sediment cores from the Eastern Equatorial Pacific were obtained and processed. The concentration  $^{60}\text{Fe}/\text{Fe}$  was then measured with accelerator mass spectrometry at the GAMS setup in Garching. It features a gas-filled magnet, allowing for complete isobar suppression in the case of  $^{60}\text{Fe}$  and  $^{60}\text{Ni}$ , leading to a sensitivity which can reach even below  $^{60}\text{Fe}/\text{Fe}=10^{-16}$ . Additionally, one of the sediment cores was also analyzed for  $^{10}\text{Be}$  and  $^{26}\text{Al}$  for independent dating of the samples at the DREAMS facility in Dresden.

**17.2 Monday 25 August - Afternoon****Topic : ISSI 01****Session 3A****Developments and applications in  $^{14}\text{C}$  gas analysis : dating a sediment core on sub-mg foraminifera samples.****Fahrni Simon<sup>1,2</sup> Wacker Lukas,<sup>1</sup> Moros Matthias,<sup>3</sup> Zillen-Snowball Lovisa.<sup>4</sup>**

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ETH Zürich has recently started to run fully automated and unattended measurements of  $^{14}\text{C}$  gas samples in cooperation with Ionplus AG. Thus, tube-sealed  $\text{CO}_2$ , carbonates and solid organics can be measured in a fully automated and highly time efficient way. This last step in automation together with high negative ion beam currents of 15 to 20  $\mu\text{A C}^-$  allow us to measure small samples containing 10 to 100 micrograms carbon with virtually no sample preparation and a precisions down to 6 ‰ on modern samples. Blank levels of 42 kyr are achieved readily. The latest developments at ETH Zurich/Ionplus AG will allow the fast analysis of hundreds of (small) samples for studies with moderate precision but high throughput. We present an application of these new ultrafast measurements on a set of benthic foraminifera (*Elphidium excavatum* spp.) samples from the central Baltic Sea. In order to establish the first high-resolution master chronology for central Baltic's Littorina Sea stage sediments, more than 30 dates were obtained for a 7 m sediment core taken at a carefully selected site. The 7 m thick sediments cover the last ca. 7600 years of the Baltic Sea's history. This new master chronology can be transferred to other site locations by correlation of fast-measured loss on ignition or XRF-scanning downcore profiles. The new master chronology will enable us, for the first time, to reliably calculate basin wide matter fluxes/ accumulation rates in the near future.

Topic : ISSI 02

Session 3A

## Flow separation of gas components with different axial momentum for online coupling of CO<sub>2</sub>-producing analytical instruments with Accelerator Mass Spectrometry.

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Efficient interfaces have been developed to couple the ultra-low sensitivity of AMS with bioanalytical instruments for the detection of radiocarbon as isotopic tracer [1]. Most CO<sub>2</sub>-producing interfaces combust samples under high pressure; delivering the CO<sub>2</sub> into the vacuum of the gas ion source along with high amount of other gases. Under these conditions, the ionization efficiency is small ( $\sim 1\%$ ) and can only be increased to 5-10% at lower gas flows [2, 3]. We developed a new device that separates the CO<sub>2</sub> from the excess helium carrier depending on their axial momentum. The inflow mixture of CO<sub>2</sub>/He enters the flow separator (FS) through a capillary that is set across a Tee connector and ends up inside of a 1/16 tubing, mounted at the front of the Tee. This tubing is connected to the feed-through of the ion source, creating a forward suction. A scroll pump is connected to the third outlet of the Tee to establish a backward suction that separates the low-mass components. The composition of the forward and backward flows was probed with a mass spectrometer. The FS was used for online coupling an elemental analyzer with an AMS as a first proof-of-concept of its interfacing capabilities for diverse hyphenations. We found that for a given backward pumping and capillary ID; there is an inflow onset where the CO<sub>2</sub> is only detected in the forward flow, towards the ion source, but not in the backward flow. The FS delivery efficiency is due to the high momentum flow of CO<sub>2</sub>. Computer simulations illustrated the CO<sub>2</sub> behavior before and after the onset. The FS reduced the total inflow from 180 mL/min to 1 mL/min, keeping the ion source pressure in the range of  $10^{-6}$  mbar.

[1] A. Thomas et al. Anal. Chem. 83 (2011) 9413

[2] G. Salazar et al. NIMB. 294 (2013) 300.

[3] S. Fahrni et al. NIMB. 294 (2013) 320

### Rapid Radiocarbon Analyses by Laser Ablation.

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Laser ablation (LA) is a powerful sampling technique which allows the removal of small quantities of material from a solid sample and its subsequent online analysis [1]. By focusing high intensity laser pulses on carbonate samples carbon dioxide is generated and can directly be introduced into the gas ion source (GIS) of an Accelerator Mass Spectrometer (AMS) [2]. The new technique [3] allows rapid radiocarbon analyses at high spatial resolution. This is especially useful in the case of carbonate records such as speleothems and corals as conventional measurements are laborious and the achievable spatial resolution is limited. For the direct coupling of LA with AMS a LA unit was developed consisting of an ablation cell (effective volume of approximately 0.6 ml) that ensures a rapid response to intensity changes and minimal particle deposition on the cell window and walls. This specific design leads to short measurement times and reduces cross-contamination. Furthermore, large samples (150 x 25 x 15 mm<sup>3</sup>) can be hosted by the cell and moved by precise positioning relative to the laser beam. An ArF-excimer laser ( $\lambda = 193$  nm) is guided to the sample surface, allowing for ablation at a scale of less than 100  $\mu$ m. The applicability of this sampling technique has been tested with pressed carbonate powder reference materials and marble. Furthermore, aspects such as ablation rates and carbon dioxide production as well as the blank value and cross contamination will be addressed.

[1] Koch, J. Günther, D. Applied Spectroscopy 2011, 65, 155A

[2] Ruff, M. Wacker, L. Gaggeler, H.W. Suter, M. Synal, H.A. Szidat, S. Radiocarbon 2007, 49, 307.

[3] Wacker, L. Münsterer, C. Hattendorf, B. Christl, M. Günther, D. Synal, H.A. NIM B 2013, 294, 287.



## An Interface for the Direct Coupling of Small Liquid Samples to AMS.

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Currently, most biochemical samples for  $^{14}\text{C}$ -AMS analysis at LLNL are converted to graphite through a multistep, time consuming and labor intensive process. This procedure necessitates significant human handling, reduces sensitivity and suffers from long turnaround times. A solution to these limitations is to combust and directly analyze the samples as  $\text{CO}_2$ , thus eliminating the need to reduce the samples to graphite. We have developed an on-line combustion interface, coupled to our  $\text{CO}_2$  gas-accepting ion source attached to our 1-MV BioAMS spectrometer, to enable the direct analysis of nonvolatile carbonaceous samples. Our interface is based on the moving wire technique and allows for both the analysis of discrete small samples, as well as for continuous flow applications to directly measure the output of a coupled HPLC in real time. Discrete samples containing a few 10s of nanograms of carbon and as little as 50 zmol  $^{14}\text{C}$  can be measured with a 3-5% precision in a few minutes. The dynamic range of our system spans approximately 5 orders in magnitude. Sample to sample memory is minimized by the use of fresh targets for each discrete sample or by minimizing the amount of  $^{12}\text{C}$  present in a peak generated by an HPLC containing a significant amount of  $^{14}\text{C}$ . Liquid sample AMS provides a new technology to expand our biomedical AMS program by enabling the capability to measure low-level biochemicals in extremely small samples that would otherwise be inaccessible.

Work performed at the Research Resource for Biomedical AMS, which is operated at LLNL under the auspices of the U.S. DOE under contract DE-AC52-07NA27344, and is supported by the National Institutes of Health under grant number 8P41GM103483.

Topic : GAA 26

Session 3A

**Age determination of metal types by measurement of "meok" using AMS.**

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"Zeungdoga", the national treasure No. 758 of Korea, is an ancient book printed using wood-type printing technology in AD 1239. In the last page of the book, it was noted that the book was reprinted one of an original book printed by metal-type printing technology. So, it is estimated that the metal-type printing technology was already used in Korea before AD 1239. However, the book printed by metal-types does not exist anymore in Korea. A set of metal types supposed to be used for printing "Zeungdoga" was discovered several years ago. KIGAM AMS Lab. has dedicated to measure their age to draw the fabricating year. Unfortunately, it was hard to directly apply radiocarbon age dating to measure their fabricating age because the types were made by bronze and no carbon was contained in them. In ancient Asia, a kind of black ink, called "meok" which is made by a mixture of soot of wood and glue, was widely used. And the types were also used with the meok. We collected meok from the surface of the types and measured the age, which may represent the last time when they were used. In this work, meok samples were collected from 15 metal types, and 13 ages were obtained. The youngest age was  $800 \pm 40$  yrBP, and the reasonable oldest one was 1320 yrBP. Weighted average after eliminating ages with poor statistics was  $1005 \pm 40$  yrBP. This age is older than the age of Jikji (AD 1377), which is a Buddhist document known to be the world oldest one printed by metal types so far, by 300 years, and the age of Gutenberg bible (AD 1450).

Topic : GAA 34

Session 3B

## Production of $^{41}\text{CaH}_2$ samples for AMS measurements. Application to Interplanetary Dust Particles.

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Interplanetary Dust Particles (IDPs) also called micrometeorites or cosmic spherules are small grains, generally less than a few hundred micrometers in size. Their main source is the Asteroid Belt located between Mars and Jupiter. During their flight from the Asteroid Belt to the Earth they are irradiated by solar and galactic cosmic rays and  $^{41}\text{Ca}$  and  $^{53}\text{Mn}$  are formed. Thus,  $^{41}\text{Ca}$  ( $T_{1/2}=1.03\times 10^5$  y) can be used as a unique tracer to determine the accretion rate of IDPs on Earth because there are no significant terrestrial sources for this radionuclide. Since a large fraction of the extraterrestrial matter evaporates during their entry in the atmosphere, the  $^{41}\text{Ca}$  in the IDP is released and can be measured by AMS using aerosols and/or ice samples. For that reason, 1.4 tons of melted snow has been collected at the permanent Argentinean Jubany Station, located in Antarctica. The production of  $^{41}\text{Ca}$  samples for AMS measurements can be made in two different ways : as fluorides or hydrides, depending on the expected  $^{41}\text{Ca}/^{40}\text{Ca}$  ratio in the samples. Because of the very low expected  $^{41}\text{Ca}/^{40}\text{Ca}$  ratios in IDP samples (on the order of  $10^{-15}$ ), the procedure to get  $^{41}\text{CaH}_2$  samples has been optimized at the Maier Leibnitz Laboratorium (MLL) to get a sensitivity down to  $10^{-16}$  for this radionuclide. First blank and standard measurements will be shown and the status of the AMS facility at MLL for the measurement of  $^{41}\text{Ca}$  will be presented.

Topic : CNA 02

Session 3B

## Multiple radionuclide study of a recent supernova event in deep-sea sediments with AMS.

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Long-lived radionuclides such as  $^{26}\text{Al}$ ,  $^{53}\text{Mn}$ , and  $^{60}\text{Fe}$  are generated in massive stars and ejected into space by stellar winds and explosions. If a star ends its life in a supernova (SN) explosion close to the solar system, a fraction of these elements might be deposited in terrestrial archives. Recent analysis of a ferromanganese crust [1], fossilized bacteria in deep-sea sediments [2] and lunar samples [3] evidence a  $^{60}\text{Fe}$  concentration enhancement  $\sim 2\text{-}3$  Myr ago, suggesting to originate from one or more SNe [1].

We expanded this work to a comprehensive and detailed study of a full set of SN-related radionuclides. Detailed depth profiles of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{53}\text{Mn}$  and  $^{60}\text{Fe}$  concentrations were measured at three different AMS laboratories for  $\sim 100$  individual samples from four deep-sea sediment cores from the Indian Ocean.

In contrast to our  $^{60}\text{Fe}$  data, which shows a clear signal without terrestrial background, a possible  $^{26}\text{Al}$  signal from a SN event is hidden within a non-negligible terrestrial background production. The major source of  $^{26}\text{Al}$  is spallogenic production by cosmic-rays in the Earth's atmosphere. We obtained isotope ratios  $^{26}\text{Al}/^{27}\text{Al}$  of  $\sim 10^{-14}$  with regularly  $< 10\%$  statistical uncertainty [4]. This allowed us to generate for the first time a full history of precise  $^{26}\text{Al}$  data over a time period of 2 Myr for two sediment cores revealing an unexpected smooth depth dependence. We took advantage of it and applied the  $^{26}\text{Al}/^{27}\text{Al}$  ratio as an independent and enhanced dating tool, comparable to  $^{10}\text{Be}/^9\text{Be}$  but without needing stable isotope measurements. Comparative measurements of  $^{10}\text{Be}/^9\text{Be}$  at the DREAMS and VERA facilities show a very good agreement ( $\sim 5\%$ ).

[1] Knie et al., Phys. Rev. Lett 93, 2004

[2] Ludwig et al., AMS-13

[3] Fimiani et al., LPSC 1659, 2012

[4] Feige et al., EPJWC, 63 2013

### Evidence for the deposition of interstellar material on the lunar surface.

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The enhanced deposition of  $^{60}\text{Fe}$  found in a deep ocean ferromanganese crust about  $(2.1 \pm 0.4)$  Myr ago (Knie et al. PRL 93, 171103 (2004), Fitoussi et al. PRL 101, 121101 (2008)) indicates that one or more supernova (SN) explosions occurred in the vicinity of the solar System. Due to its lack of atmosphere and its negligible sedimentation rate, the Lunar surface is an excellent quantitative reservoir for SN debris. We searched for live  $^{60}\text{Fe}$  (half life 2.62 Myr) and  $^{53}\text{Mn}$  (half life 3.7 Myr) in different samples from 3 Apollo missions.  $^{53}\text{Mn}$  is, similar as  $^{26}\text{Al}$  and  $^{60}\text{Fe}$ , a tool to trace nucleosynthesis activities. It is formed primarily during the explosive silicon-burning of the inner shells of SNe via  $^{53}\text{Fe}$  which beta-decays to  $^{53}\text{Mn}$ . Samples where we found an enhanced  $^{60}\text{Fe}$  concentration showed also an enhancement of  $^{53}\text{Mn}$ . This could be the first detection of live  $^{53}\text{Mn}$  originating from recent nucleosynthesis. The measurements were performed at the Maier - Leibnitz Laboratorium in Munich, Germany. With the Gas-filled Analyzing Magnet System (GAMS) concentrations of  $^{60}\text{Fe}/\text{Fe}$  down to  $10^{-16}$  or even below can be measured.

Topic : NFF 08

Session 3B

**A dedicated AMS setup for  $^{53}\text{Mn}/^{60}\text{Fe}$  at the Cologne FN Tandem Accelerator.**

Schiffer Markus,<sup>1</sup> Dewald Alfred,<sup>1</sup> Feuerstein Claus,<sup>1</sup> Altenkirch Richard,<sup>1</sup> Stolz Alexander,<sup>1</sup> Heinze Stefan,<sup>1</sup>

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Following demands for AMS measurements of medium mass isotopes, especially for  $^{53}\text{Mn}$  or  $^{60}\text{Fe}$  we started to build a dedicated AMS setup at the Cologne FN Tandem accelerator . This accelerator with a maximum terminal voltage of 10 MV can be reliably operated at a terminal voltage of 9.5 MV which corresponds to energies of 95-104.5 MeV for  $^{60}\text{Fe}$  or  $^{53}\text{Mn}$  beams using the 9+ or 10+ charge state. These charge states can be obtained with foil stripping with efficiencies of 30% and 20%, respectively. Energies around 100 MeV will be sufficient to effectively suppress the stable isobars  $^{60}\text{Ni}$  and  $^{53}\text{Cr}$  by (dE/dx) techniques using combinations of energy degrader foils and dispersive elements like electrostatic analyzers or gas filled magnets as well as (dE/dx) ion detectors. Alternatively a time of flight (TOF) system can be used. A low energy mass spectrometer has been built, consisting of a NEC multi-cathode-sputter ion source, a spherical ESA and a double focusing  $90^\circ$  magnet. This mass spectrometer became operational recently and first beams have been successfully injected into the FN tandem accelerator. The high energy mass spectrometer has been designed and the main components are available or have been ordered. Some components of the low and high energy mass spectrometer are stemming from the AMS facility at Utrecht which has been closed 2009. It is scheduled to have the high energy mass spectrometer finished in spring 2015. In this contribution we will report on details of the complete setup and on the expected features of the new AMS setup which is based partly on measurements and on calculations.

Topic : AAT 21

Session 3B

**Application of AMS for the Analysis of Primordial Nuclides in High Purity Copper.**

**Famulok Nicolai,<sup>1</sup> Hain Karin,<sup>1</sup> Faestermann Thomas,<sup>1</sup> Fimiani Leticia,<sup>1</sup> Gómez Guzmán José Manuel,<sup>1</sup> Korschinek Gunther,<sup>1</sup> Ludwig Peter,<sup>1</sup> Schönert Stefan.<sup>1</sup>**

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The sensitivity of experiments in rare event physics like neutrino or direct dark matter detection crucially depends on the background level. Therefore, all material surrounding the detectors requires low contamination of radionuclides in order not to create additional background. A significant contribution originates from the primordial actinides thorium and uranium and the progenies of their decay chains. At the Maier Leibnitz Laboratorium in Munich the applicability of ultra-sensitive Accelerator Mass Spectrometry (AMS) for the direct detection of thorium and uranium impurities in a copper matrix was tested for the first time. For this special purpose, Th and U were extracted from the ion source as a copper compound. Different samples of copper, high purity copper and of a copper alloy were investigated. The lowest concentrations achieved in AMS measurements until now were  $(1.4 \pm 0.6) \cdot 10^{-11}$  g/g for thorium and  $(7 \pm 4) \cdot 10^{-14}$  g/g for uranium which correspond to  $(56 \pm 16) \mu\text{Bq/kg}$  and  $(0.9 \pm 0.5) \mu\text{Bq/kg}$ , respectively. The particular requirements on the AMS technique and the developed measurement procedure will be presented, followed by a discussion of the results of the first measurements.



Topic : SP 11

Session 4A

## Progress in automated extraction and purification of in situ $^{14}\text{C}$ from quartz : results from the Purdue in situ $^{14}\text{C}$ laboratory.

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Current extraction methods for in situ  $^{14}\text{C}$  from quartz (e.g. Lifton et al. 2001 ; Pigati et al. 2010 ; Hippe et al. 2013) are time-consuming and repetitive, making them an attractive target for automation. We report on the status of in situ  $^{14}\text{C}$  extraction and purification systems originally automated at the University of Arizona that have now been reconstructed and upgraded at PRIME Lab. The Purdue in situ  $^{14}\text{C}$  laboratory builds on the flow-through extraction system design of Pigati et al. (2010), automating most of the procedure by retrofitting existing valves with external servo-controlled actuators, regulating the pressure of research purity  $\text{O}_2$  inside the furnace tube via a PID-based pressure controller in concert with an inlet mass flow controller, and installing an automated liquid  $\text{N}_2$  distribution system, all driven by LabView® software. A separate system for cryogenic  $\text{CO}_2$  purification, dilution, and splitting is also fully automated, ensuring a highly repeatable process regardless of the operator. We will present results from blanks and intercomparison material (CRONUS-A), as well as results of experiments to increase the amount of material used in extraction, from the standard 5 g to 10 g or above.

Hippe, K. Kober, F. Wacker, L. Fahrni, S.M. Ivy-Ochs, S. Akcar, N. Schluchter, C. Wieler, R. 2013. Nuclear Inst. and Methods in Physics Research, B 294, 81-86.

Lifton, N. Jull, A. Quade, J. 2001. Geochimica et Cosmochimica Acta 65, 1953-1969.

Pigati, J. Lifton, N. Jull, A. Quade, J. 2010. Radiocarbon 52, 1236-1243.

Topic : CRI 01

Session 4A

### Improving the tree-based radiocarbon calibration curve

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Trees catalogued by dendrochronology are the most valuable source for  $^{14}\text{C}$  calibration, as past fluctuations of the atmospheric  $^{14}\text{C}$  level are preserved in tree rings. The longest European chronology, reaching back to 12,594 BP, has been built on oak and pine collected in gravel pits from the river valleys of Main, Danube and Rhine in southern Germany with a short extension on pine chronologies built from finds in the Zurich area. While the  $^{14}\text{C}$  is preserved with annual resolution in trees, the major part of the IntCal13 calibration curve is presently based on measurements on decadal samples. We will demonstrate on examples that a higher temporal resolution would be beneficial for more precise radiocarbon dating. We will also show that a significant extension of the dendrochronologically based calibration curve of about 2000 years into the late glacial times may soon be realized. A total of 270 well-preserved pine trunks were found in 2013 in the town of Zürich and were pre-dated with radiocarbon to cover a time range of about 11,500 BP to 14,400 BP.

Topic : FNS 3

Session 4A

**Age estimation based on an annual variation curve of bomb-produced radiocarbon concentration : growth process of an elephant tusk.**

Nakamura Toshio,<sup>1</sup> Koike Hiroko,<sup>2</sup> Aizawa Jun,<sup>3</sup> Mitsuru Okuno.<sup>3</sup>

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[2]Kyushu University Museum, (Japan)

[3]Faculty of Science, Fukuoka University, (Japan)

Atmospheric concentration of radiocarbon ( $^{14}\text{C}$ ) produced by atmospheric nuclear bomb tests from 1945 to 1963, shows a unique secular variations from 1955 to present, slightly dependent on spatial locations (nuclear test sites), it is possible to be used for age estimation of natural carbonaceous materials in the time range with great success. In East Asian countries, we are normally accustomed to own personal seal made of animal tusk, stone, wood, etc. and one of the excellent source materials is considered as ivory. In 1973 the Washington Convention (CITES) was organized and signed worldly to prohibit the trading of endangered species of wild fauna and flora, living, dead, or products, and of course elephant tusk is one of them. Since the CITES was signed by Japan in 1980, elephant tusk or its products permitted for trading are those that were already owned personally or officially before 1980. Thus it is important to check by  $^{14}\text{C}$  concentration whether the ivory is from an elephant that was dead before 1980, for trading purposes, if there exists no official document that proves that they were produced before 1980. A full elephant tusk with 175 cm long and 13.8 cm diameter at the tusk root owned by the Kyushu University Museum was offered for  $^{14}\text{C}$  analysis. We selected parts of tusk samples, and by  $^{14}\text{C}$  analysis we analyzed formation ages of each parts. The  $^{14}\text{C}$  concentration of tusk roots suggested that the elephant was estimated to be dead at around 1994.

Topic : GAA 03

Session 4A

## Compound specific radiocarbon analysis from indoor air samples via accelerator mass spectrometry.

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Many volatile organic environmental compounds are potentially dangerous due to their allergic or carcinogen impact on humans. For the establishment of effective countermeasures for lowering their concentration in houses, sources have to be known. Our investigation is focused on aldehyde compounds since their indoor concentration is often above the official guidelines and since they originate from biogenic or anthropogenic sources. Both types of sources can be distinguished by their different  $^{14}\text{C}$  content which can be measured via accelerator mass spectrometry (AMS). For the collection and separation of these gaseous substances they have to be converted into liquid or solid phase by derivatization. This leads to the incorporation of up to six additional carbon atoms into the derivatized sample and hence to a reduced  $^{14}\text{C}$  content. To reduce the number of additional carbon atoms and to optimize efficiency and duration of the procedure, different derivatization compounds and methods have been tested with acet- and formaldehyde of known  $^{14}\text{C}$  content. The Erlangen AMS facility, based on an EN tandem accelerator and a hybrid sputter ion source for solid and gaseous samples, is well suited for the measurement of isotope ratios  $^{14}\text{C}/^{12}\text{C} \approx 10^{-12}$  -  $10^{-15}$ . The  $^{14}\text{C}$  concentration of the calibration samples and from indoor air samples in apartments, beer taverns and schools have been determined by AMS, the corresponding results are discussed with regard to potential sources of aldehydes.

Topic : CNA 09

Session 4B

**Aeolian soil loss from agricultural areas as monitored by fallout plutonium****Dunai Tibor J.,<sup>1</sup> Wiesel H.<sup>1</sup> Amelung W.<sup>2</sup> Dewald A.<sup>3</sup> Feuerstein C.<sup>3</sup> Fifield L.K.<sup>4</sup> Tims S. G.<sup>4</sup> Heinze S.<sup>3</sup>**

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[3]CologneAMS/Institute of Nuclear Physics, University of Cologne (Germany)

[4]Australian National University (ANU), (Australia)

Soils from three agrosystems in the Free State Province, South Africa were studied to quantify soil-loss occurring during the 50 years following the input of fallout plutonium from atmospheric nuclear weapons testing. The investigated areas are characterized by low gradients ( $< 2^\circ$ ) and are cultivated for cereal production. For each sub-area within the agrosystem the timing of the transformation from savanna to agricultural ground is known, ranging from 90 years to less than 5 years. Remaining near pristine savanna areas were used as reference areas to gauge local fallout nuclide input. Using  $^{242}\text{Pu}$  as spike, we measured the remaining activity of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  (AMS). A subset of samples was also analyzed for  $^{137}\text{Cs}$  ( $\gamma$ -ray spectrometry). The  $^{240}\text{Pu}/^{239}\text{Pu}$  of all samples are indistinguishable from southern Hemisphere atmospheric fallout. Likewise, the uniform  $^{137}\text{Cs}/^{239}\text{Pu}$ -ratios in all measured samples reflect their common fallout origin. Furthermore, the uniform  $^{137}\text{Cs}/^{239}\text{Pu}$ -ratio signifies that both species are retained in the same soil fraction(s). We observe for all three agrosystems that during the first 20 years of cultivation nearly 50 % of the fallout plutonium inventory is removed. Since plutonium and caesium bind predominantly on small particle sizes, the loss of fallout inventory reflects the loss of the fertile fine-grained soil fractions. It is likely that the soil loss is largely due to wind erosion, since the low gradients are not amenable to efficient fluvial erosion.

Topic : CRI 03

Session 4B

## Towards high precision and low ratio $^{10}\text{Be}$ measurements with the SUERC 5MV tandem : bigger isn't always better

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Miguéns-Rodríguez Maria,<sup>1</sup> Bierman Paul,<sup>3</sup> Carlson Anders,<sup>4</sup> Freeman Stewart.<sup>1</sup>

[1]Scottish Universities Environmental Research Centre, (United Kingdom)

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We quantify the uncertainties, total system efficiency, and interlaboratory reproducibility of  $^{10}\text{Be}$  measurements on the SUERC 5MV spectrometer. Secondary standards have average statistical uncertainties based on counting statistics of 0.6%, 1.0%, and 1.3% for standard materials with ratios of  $3 \times 10^{-12}$  (N=144),  $1 \times 10^{-12}$  (N=86), and  $6 \times 10^{-13}$  (N=81), respectively. The average measured ratios fall within the reported 1.1% uncertainty. The error-weighted standard deviation is 1.5%, 2.6%, and 2.6%, respectively. These data indicate an additional uncertainty of 1.4-2.4% above counting statistics. Furthermore, we measured 11 replicate quartz targets. These full-chemistry replicates have an average uncertainty of 1.2%, but a standard deviation of 3.6%. We also determined the minimum ionization and total system efficiency to be 0.3% and 0.1%, respectively.  $^{10}\text{Be}$  samples prepared by chemistry laboratories in the US and measured at SUERC had high and consistent beam currents,  $105 \pm 5\%$  (1 SD) of the primary standards, indicating the high quality of chemistry (high  $\sim 100\%$  Be yield and pure BeO as verified by ICP-OES data on final Be fractions). The consistently low carrier blanks ( $6 \times 10^{-16}$ ) demonstrate the sensitivity of the SUERC system. Measured 1-sigma analytical uncertainties were 3% for low  $10^{-13}$ , 5-12% for  $10^{-14}$ , and 14-19% for low  $10^{-15}$  ratio samples. An AMS laboratory intercomparison of 3 quartz samples ranging from  $\sim 3 \times 10^{-14}$  to  $\sim 1 \times 10^{-13}$  are reproducible within their reported uncertainties. In the same experiments, we measured exposure ages of about  $1.4 \text{ ka} \pm 100\text{-}200$  years. These results demonstrate the effectiveness of the SUERC 5MV spectrometer, including potential for high precision and low ratio  $^{10}\text{Be}$  analyses.

Topic : GAA 01

Session 4B

**Evidence for cosmic-ray spike at 774/5 AD in annual resolution  $^{10}\text{Be}$  record of West Antarctic ice core.****Welten Kees,<sup>1</sup> Nishiizumi Kuniyuki,<sup>1</sup> Woodruff Thomas,<sup>2</sup> Caffee Marc.<sup>2</sup>**

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The annual  $^{14}\text{C}$  record in tree-rings shows evidence for a sharp spike in the cosmic-ray flux at 774/5 AD (Miyake et al. 2012). Although the cause of this cosmic-ray spike is still a topic of debate, with supernova, short gamma-ray bursts and solar flares as possible candidates, it should stand out in the annual  $^{10}\text{Be}$  record of ice cores. To better constrain the magnitude and duration of the 774/5 AD event, we measured  $^{10}\text{Be}$  concentrations in annual layers of the West Antarctic Ice Sheet (WAIS) Divide core from 295-305 m depth, corresponding to preliminary ages of  $\sim 760$ -800 AD. While most samples in this depth interval show  $^{10}\text{Be}$  concentrations of  $(10\text{-}19) \times 10^3$  atoms/g, three annual between 302.7-303.5 m depth show elevated  $^{10}\text{Be}$  concentrations up to  $35 \times 10^3$  atoms/g. This spike is 50% larger than any other event in the annual WAIS Divide  $^{10}\text{Be}$  record of the last 400 years, and the location of the spike is within uncertainty (of  $\pm 7$  yr) consistent with the spike of 774/5 AD found in the  $^{14}\text{C}$  record. The  $^{10}\text{Be}$  spike at 774/5 AD can be explained by a short-term ( $\sim 1$  year) increase in cosmic-ray flux by a factor of 5-6 and an atmospheric residence time of  $\sim 1$  year. The increase in the  $^{10}\text{Be}$  and  $^{14}\text{C}$  production rate around 774/5 AD is consistent with a solar flare similar in hardness but with a 40-50 times higher flux than the February 1956 solar flare. If so, this solar flare was the largest in the last 2000 years and provides an excellent chronological marker for ice core research.

Topic : GAA 40

Session 4B

## Long-term hillslope erosion rates of Yakushima Island, southern Japan deduced from cosmogenic $^{10}\text{Be}$ in river sediments

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Yakushima Island, southern Japan is a non-volcanic island, which mainly consists of granite. Modern precipitation rates in Yakushima range from 2400-7400 mm/yr, which is one of the highest one in Japan. This study investigated relationships between hillslope erosion rates of Yakushima Island and factors such as precipitation, basin slope, altitudes and so forth. We present new basin-averaged erosion rates from cosmogenic  $^{10}\text{Be}$  in quartz grains from 10 river sediments in Yakushima Island. We found that erosion rates of Yakushima Island are relatively low within a similar range to those in regions of several times lower rainfall than Yakushima Island, suggesting that rainfall is not necessarily a main factor of hillslope erosion. The landform evolution in Yakushima Island is characterized by that hillslopes are eroded very slowly over the timescale of the order of  $10^3$ - $10^4$  yrs even though Yakushima has extraordinary much amount of rainfall. In this presentation, we focus on a pattern of hillslope erosion and a process of landform evolution in Yakushima Island, compared to erosion manners of other granitic mountains in Japan which vary in tectonic and climatic settings.



**17.3 Tuesday 26 August - Morning****Topic : AAT 15****Session 5****Progress in design and performance of low energy AMS systems.**

Synal Hans-Arno,<sup>1</sup> Christl Marcus,<sup>1</sup> Maxeiner Sascha,<sup>1</sup> Müller Arnold,<sup>1</sup> Seiler Martin,<sup>1</sup> Wacker Lukas.<sup>1</sup>

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AMS systems based on vacuum insulated accelerators (MICADAS) can be regarded as versatile radiocarbon instruments enabling superior conditions, in respect of measurement performance and sample throughput. At present there are nine MICADAS instruments in operation worldwide. With the production of new MICADAS systems, we have always launched a continuous development program to increase system performance. Beside the progress with the gas interface components, which will be reported elsewhere, significant technical improvements have been made. The instruments are now incorporating He gas stripping resulting in beam transmissions of nearly 50% and due to reduced optical beam losses in even more stable measurement conditions. The latest version of a MICADAS system is fully equipped with fixed field magnets reducing significantly operation costs of the instruments. Furthermore, we report on tests to demonstrate that the MICADAS concept is not limited to radiocarbon detection, only. By increasing the terminal voltage to 300kV higher beam energies become possible allowing detection of very heavy nuclides such as actinides or  $^{129}\text{I}$  and to a limited extend  $^{10}\text{Be}$  and  $^{26}\text{Al}$ .

**$^{26}\text{Al}$  measurements below 500 kV.**

Müller Arnold Milenko,<sup>1</sup> Christl Marcus,<sup>1</sup> Lachner Johannes,<sup>1</sup> Maxeiner Sascha,<sup>1</sup> Synal Hans-Arno,<sup>1</sup>  
Zanella Claudia.<sup>1</sup>

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For the majority of radionuclides measured at compact AMS facilities the use of helium as stripper gas improved the measurement efficiency considerably. Prominent examples are  $^{14}\text{C}$ ,  $^{10}\text{Be}$ ,  $^{129}\text{I}$  and the actinides [1,2,3]. Helium stripping would be also an attractive option for measurements of  $^{26}\text{Al}$  at terminal voltages around 500 kV since transmissions of about 55 % for the 2+ charge state were achieved. However, at the 2+ charge state high  $^{13}\text{C}^+$  intensities originating from the  $^{13}\text{C}_2$  breakup make background suppression difficult. An approach was tested by introducing a passive absorber in front of the detector in order to stop the  $^{13}\text{C}_2$  breakup in the absorber material. Initial results obtained at the ETH TANDY facility using a very simple absorber design were promising [3]. These findings motivated the design of a new gas ionization chamber equipped with a separate gas absorber cell mounted in front of the detector. The absorber is enclosed by two SiN foils. First measurements with the new detector-absorber configuration were performed at 500 kV and 300 kV at the ETH Zurich TANDY facility. For  $^{26}\text{Al}$  an overall transmission from the low energy side into the detector of more than 40 % at 500 kV and of about 25 % at 300 kV were achieved. Various blank materials were measured at the level  $1 \times 10^{-13}$  and below. The suppression and stopping of  $^{13}\text{C}_2$  in the absorber were investigated systematically. The results will be discussed and compared to SRIM [4] simulations in this presentation.

[1] Schulze-König et al. NIM B 269 (2011) 34-39

[2] Vockenhuber et al. NIM B 294 (2013) 382-386

[3] Lachner et al. NIM B In Press (2014)

[4] Ziegler et al. NIM B 268 (2010) 1818-1823

Topic : AAT 14

Session 5

**Further improvement for  $^{10}\text{Be}$  measurements with an upgraded compact radiocarbon facility.****Fu Dongpo,<sup>1</sup> Ding Xingfang,<sup>1</sup> Liu Kexin,<sup>1</sup> Müller Arnord,<sup>2</sup> Suter Martin,<sup>2</sup> Zhou Liping,<sup>3</sup> Synal Hans-Arno.<sup>1</sup>**

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In the first stage of the attempt to upgrade the Peking University 500kV NEC radiocarbon facility for  $^{10}\text{Be}$  measurement, we mounted a silicon nitride foil as the secondary stripper in front of the electrostatic deflector and used an ETHZ-designed high-resolution Delta E-Eres gas ionization chamber to replace the original Si detector (for radiocarbon measurement). This simple arrangement has yielded a  $^{10}\text{Be}/^9\text{Be}$  background level as low as  $3.4 \times 10^{-14}$  [1]. Recently, we have installed a  $90^\circ$  magnet after the electrostatic deflector, which is expected to further reduce the background and increase the transmission by re-focusing the beryllium ions. The silicon detector will be shifted slightly relative to its original position but can be lifted up manually when  $^{10}\text{Be}$  measurement is carried out. The gas detector for  $^{10}\text{Be}$  is mounted at the end of the beam line after the new magnet. The lay-out which was chosen is very compact and does not require more space than the original instrument. We shall report the performance of  $^{10}\text{Be}$  measurement with the new arrangement.

[1] A M Müller et al. (2013) Radiocarbon, 55 (2-3), 231-236.

## Memory effects using an elemental analyser to combust radiocarbon samples : failure and recovery.

Fedi M.E.,<sup>1</sup> Liccioli L.,<sup>1,2</sup> Castelli L.,<sup>1</sup> Czelusniak C.,<sup>1,3</sup> Giuntini L.,<sup>1,3</sup> Mandò P.A.,<sup>1,3</sup> Palla L.,<sup>4</sup>  
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In the combustion and graphitization line for  $^{14}\text{C}$ -AMS samples used at INFN-LABEC for archaeological and geological applications, samples are burnt using an elemental analyser (EA). Advantages and drawbacks of EAs are known, a drawback being the possibility to introduce some contaminations or memory effects. Different parts inside an EA, e.g. the autosampler and the gas-chromatography column, might in principle be responsible of such problems. During a measurement run some time ago, we measured, indeed, radiocarbon concentration values somewhat higher than usual in nominally blank samples. These "bad" data were explainable by memory effects and we were able to apply appropriate corrections by assuming that each measured radiocarbon apparent concentration was altered by the contamination of a given amount of the sample burnt immediately before : indeed, by repeating cycles of sequential combustions of standards and blanks, we observed a good reproducibility of the amount of contamination from the previous sample needed to explain the results. However, we were obviously unhappy with the fact itself of such corrections being needed, and several tests were performed to identify the source of contamination and eliminate it. Eventually, we succeeded in finding the cause of this failure and in recovering the full efficiency of the system. Here we report about our experience.

**Systematic investigation on contamination of background AMS samples.**

Paul Dipayan,<sup>1</sup> Been Henk A.<sup>1</sup> Th. Aerts-Bijma Anita,<sup>1</sup> Meijer Harro A. J.<sup>1</sup>

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Accelerator Mass Spectrometric (AMS) measurements of radiocarbon content in very old samples are often challenging and carry large relative uncertainties due to possible contaminations coming from the natural surroundings. In case of very old samples, the natural surrounding levels of radiocarbon are 2-3 orders of magnitude higher than the samples themselves. Hence, precautions are involved during the preparation steps to have the samples pristine till measurements are performed. As samples frequently have to be temporarily stored until AMS measurements can be performed, storage conditions form a crucial part of these precautions. Here we describe a systematic assessment of the process of contamination of graphite from very old AMS samples with alien  $^{14}\text{C}$  from the surroundings. Samples, pressed graphite (on AMS targets) and graphite powder, were stored in various storage conditions to identify and catalogue the conditions that effected the most. Storage conditions included elevated levels of modern  $\text{CO}_2$ , to accelerate the process of uptake leading to noticeable effects. Previous experiments and the ongoing ones have clearly shown that the pressed targets are more vulnerable to contamination than the unpressed graphite, stored in similar conditions. All samples, after fixed storage duration, were measured at the Groningen AMS facility. This assessment aims to provide a better insight on minimizing the contamination and identifying the steps and mechanism of the introduction of alien  $^{14}\text{C}$ .

## Identification of Actinides Emitted into the Pacific Ocean by the Fukushima Accident

**Hain Karin,<sup>1</sup> Faestermann Thomas,<sup>1</sup> Famulok Nicolai,<sup>1</sup> Fimiani Leticia,<sup>1</sup> Gómez Guzmán José Manuel,<sup>1</sup> Korschinek Gunther,<sup>1</sup> Kortmann Florian,<sup>1</sup> Lierse Von Gostomski Christoph,<sup>1</sup> Ludwig Peter.<sup>1</sup>**

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In order to obtain an estimate on the entry of actinides into the Pacific Ocean due to the Fukushima accident, we want to determine in the sea water the isotopic ratios of plutonium,  $^{241}\text{Pu}/^{240}\text{Pu}$  and  $^{240}\text{Pu}/^{239}\text{Pu}$ , which are unique for the Fukushima Nuclear Power Plant (NPP) compared to nuclear fallout. In addition, we are also interested in the inventory and the distribution of Pu, Np and Am in the Pacific Ocean. For this purpose we are going to use sea water samples taken at different locations and depths in the Pacific Ocean.

At first, a chemical procedure for actinide separation had to be developed at the Maier-Leibnitz-Laboratory (MLL) in Munich which matches the special demands of AMS and of our lab. One of the major goals was to obtain a highly effective chemical separation of  $^{241}\text{Pu}$  and its daughter nuclide  $^{241}\text{Am}$  and a reproducible chemical yield of Np. The most convenient option to meet these requirements seemed to be extraction chromatography using the resins from Eichrom Technologies. In a set of experiments at the Radiochemistry Department the procedure was optimized such that the Pu and Np fractions are obtained with high chemical recovery, which was monitored by alpha- and gamma-spectroscopy. In parallel, those samples were used to improve the separation ability of neighboring masses of the AMS setup and to determine the respective blank levels.

A short motivation for the AMS measurements of actinides in the Pacific Ocean relating to the Fukushima accident will be given and the chemical separation method will be presented. First results of measurements of blank and ocean water samples will be discussed, followed by an outlook on the remaining steps to the identification of a Fukushima NPP entry.

Topic : FNS 2

Session 6A

## **Iodine-129 and other radionuclides in the atmosphere at Tsukuba, Japan, after the Fukushima Daiichi Nuclear Power Plant accident.**

**Shibata Yasuyuki,<sup>1</sup> Kato Humiaki,<sup>1</sup> Kobayashi Toshiyuki,<sup>1</sup> Yamakawa Akane,<sup>1</sup> Doi Taeko,<sup>1</sup> Tanaka Atsushi,<sup>1</sup> Xu Sheng,<sup>2</sup> Freeman Stewart,<sup>2</sup> Masumoto Kazuyoshi,<sup>3</sup> Toyoda Akihiro.<sup>3</sup>**

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[2]Scottish Universities Environmental Research Centre (United Kingdom)

[3]High Energy Accelerator Organization (Japan)

After the nuclear power plant accident in Fukushima caused by the Great East Japan earthquake and subsequent tsunami, a large amount of radionuclides was emitted into the environment. We started air sampling from the afternoon of 15th March, 2011, at Tsukuba, c.a. 170 km distant from the damaged power plant, and analyzed gamma-ray emitting nuclides in both particulate and gaseous fractions (Doi et al. (2013) J. Environ. Radioact. 122, 55). Here we report the results of iodine-129 analysis of the filters employed by accelerator mass spectrometers at NIES-TERRA and SUERC; iodine-129 in the charcoal filter was efficiently extracted by tetramethylammonium hydroxide. Iodine-129/iodine-131 atomic ratios were calculated to be around 20 on average, similar to the reported values from soil (Miyake et al. (2012)) and rain (Xu et al. (2013)) analysis. Detailed analytical procedures and data together with other radionuclides levels will be reported.

Topic : GAA 11

Session 6A

## Study on the iodine transportation in the fluvial system by analysis of iodine-129 concentration in water samples collected around the Fukushima Daiichi Nuclear Power Plant

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According to Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, vast amount of radioactive nuclides including radioactive iodine were spilled out into the environment. A rare isotope Iodine-129 ( $^{129}\text{I}$ ) was also widely distributed in a very short time by the FDNPP accident.  $^{129}\text{I}$  directly landing on the soil surface had been trapped in the upper layer of the soil and the depth profile should indicate the migration and the interaction with the soil. If  $^{129}\text{I}$  was trapped in the woods, it seems to take rather longer time for landing on the ground. Either way, a certain portion of the  $^{129}\text{I}$  should be moving downward and finally washed out by the groundwater or river with a certain rate and transported into the sea. The concentration of  $^{129}\text{I}$  in environmental water samples taken from rivers and ponds are considered to reflect the iodine transportation process by the fluvial system. In this study,  $^{129}\text{I}$  concentration of river waters were collected from Iitate village and Minami-Soma city (North to North-west of FDNPP) and  $^{129}\text{I}$  concentrations were measured by AMS. The results showed as high as  $1.0 \times 10^9$  atoms/L and had not vary significantly during period from March to October, 2012. This concentration is quite high compared to the pre-accident level ( $1\text{--}2 \times 10^6$  atoms/L), which was determined from the result of measurement for tap water collected in 2006. The combination of  $^{129}\text{I}/^{127}\text{I}$  ratio and  $^{127}\text{I}$  concentration indicates 3 sources : fossil rain water (ground water), the rain radioactively contaminated by FDNPP accident, and the iodine dissolved from soil.



## Depth profiles of $^{129}\text{I}$ and $^{129}\text{I} / ^{127}\text{I}$ ratio in soil at the near-field site of the Fukushima Dai-ichi Nuclear Power Plant.

Matsunaka Tetsuya,<sup>1</sup> Sasa Kimikazu,<sup>1</sup> Sueki Keisuke,<sup>1</sup> Takahashi Tsutomu,<sup>1</sup> Matsumura Masumi,<sup>1</sup>  
Satou Yukihiro,<sup>1</sup> Shibayama Nao,<sup>1</sup> Kitagawa Jun-Ichi,<sup>2</sup> Kinoshita Norikazu,<sup>3</sup> Matsuzaki Hiroyuki.<sup>4</sup>

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Massive nuclear fission products such as radioiodine were deposited on the surface in Fukushima via radioactive plume derived from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. In order to evaluate inventory and penetration of accident-derived  $^{129}\text{I}$  in the highly-contaminated area, depth profiles of  $^{129}\text{I}$  concentration and  $^{129}\text{I}/^{127}\text{I}$  ratio in 5cm-long soil cores after the accident were investigated at three sites : NM-6 site at Namie town (7.5 km northwest site from the FDNPP), Iw-2 site at Okuma town (4.3 km west site from the FDNPP) and Iw-8 site at Okuma town (8.2 km west site from the FDNPP). Total  $^{129}\text{I}$  inventories in the soil cores were estimated to be  $1.80 \text{ Bqm}^{-2}$  at NM-6,  $1.84 \text{ Bqm}^{-2}$  at Iw-2 and  $0.68 \text{ Bqm}^{-2}$  at Iw-8. Average  $^{129}\text{I} / ^{127}\text{I}$  ratio in each soil cores after the accident (NM-6 :  $8.6 \times 10^{-7}$ , Iw-2 :  $2.1 \times 10^{-6}$ , Iw-8 :  $4.2 \times 10^{-7}$ ) shows typical ratios of the contaminated surface soils in Fukushima. Therefore, accident-derived  $^{129}\text{I}$  deposited at NM-6 and Iw-2 were 2.7 times higher than that at Iw-8. Depth profiles of  $^{129}\text{I}$  concentration and  $^{129}\text{I} / ^{127}\text{I}$  ratio exponentially declined with depth. Approximately 90% of deposited  $^{129}\text{I}$  in 5-cm-long soil cores were observed in the surface layer of  $16.1 - 17.8 \text{ kgm}^{-2}$  (1.6 - 2.5 cm) in depth. In addition, the relaxation mass depths ( $h_0$ ) of  $^{129}\text{I}$  were determined to be  $8.2 \text{ kgm}^{-2}$  at NM-6,  $9.5 \text{ kgm}^{-2}$  at Iw-2 and  $9.2 \text{ kgm}^{-2}$  at Iw-8.

Topic : GAA 36

Session 6A

## Speciation analysis of Iodine-127 and 129 in the soil contaminated by the Fukushima Dai-ich Nuclear Power Plant accident.

Honda Maki,<sup>1</sup> Hiroyuki Matsuzaki,<sup>2</sup> Saitou Takumi,<sup>3</sup> Nagai Hisao.<sup>4</sup>

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In previous study, we investigated the depth profiles of the Fukushima derived Iodine-129 and downward migration speed in a crop filed soils near the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). It was found that  $^{129}\text{I}$  typically showed the highest concentration in the surface layer and sudden decreasing trend with depth. From the time course observation of the depth profile at the same crop field, the  $^{129}\text{I}$  downward migration rate was estimated to be ca.  $0.5 \text{ g cm}^{-2} \text{ yr}^{-1}$  which was higher than that of  $^{137}\text{Cs}$  (ca.  $0.3 \text{ g cm}^{-2} \text{ yr}^{-1}$ ). To elucidate the migration mechanism of  $^{129}\text{I}$  in soils, the speciation analyses of both isotope  $^{127}\text{I}$ ,  $^{129}\text{I}$  were conducted. The sequential extraction method, that had been developed for the analysis of  $^{137}\text{Cs}$ , was modified for the iodine extraction. A specific feature of our method is using Tetramethylammonium hydroxide as the eluent and the dialysis to separate the permeate from the retentate. In the permeate, the iodine released from organic matter is considered to be included. In our method, a soil sample is divided into 5 fractions, i.e. Water soluble (WS), Exchangeable (EX), Oxide-bound (OX), Organic matter-bound fraction (OM) and Residue (RS). The soil sample treated in this study was Ando soil taken from a crop field in Iitate Village, one of severely contaminated areas by the FDNPP accident.  $^{127}\text{I}$  concentration was measured by ICP-MS and  $^{129}\text{I}$  was determined by AMS with carrier. The distribution of  $^{127}\text{I}$ ,  $^{129}\text{I}$  in each fraction is similar, OX + OM is dominant. With close observation,  $^{129}\text{I}$  is distributed a little more in WS + EX than  $^{127}\text{I}$ . The relation of the results of speciation analysis and the migration mechanism for  $^{127}\text{I}$  and/or  $^{129}\text{I}$  in soil will be discussed.

Topic : NFF 03

Session 6B

**Status of the "new" AMS facility in Trondheim.**

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[2]Department of Archaeometry, Norwegian University of Science and Technology, (Norway)

[3]High Voltage Engineering Europa B.V, (Netherlands)

The Radiocarbon laboratory of the Norwegian University of Science and Technology in Trondheim has a long history dating back to the 1950s. Its relatively new AMS facility is based on a 1 MV Tandetron from High Voltage Engineering Europa B.V. that is equipped with a hybrid solid/gas SO-110 ion source, a low energy spectrometer supporting sequential injection, a high energy analysis system consisting of a magnet and an electrostatic deflector, allowing insertion of an absorber foil for isobar suppression and a two dimensional gas ionization detector (E and  $\Delta E$ ). The system is at present capable of measuring  $^{10}\text{Be}$ ,  $^{14}\text{C}$  and  $^{26}\text{Al}$  and can be easily modified to measure isotopes of higher masses. Our first priority is the understanding of the ion optical properties of the system for carbon isotopes. This should point the way to possible improvements in  $^{14}\text{C}$  background and measurement efficiency and optimize the system for radiocarbon measurements - also for very small samples - as a research and service facility. A further priority is an ion-optical feasibility study of the AMS measurement of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and other, heavier isotopes. The performance of the system for  $^{14}\text{C}$  measurements using the +2 charge state as well as the +1 and +3 charge states will be presented. Measuring  $^{14}\text{C}$  using the charge state +2 after acceleration can introduce a contribution from the  $^7\text{Li}_2^-$  molecule to the overall machine background. Even if the system has demonstrated a very low background (70 000  $^{14}\text{C}$  years BP or  $2 \times 10^{-16}$  on Alfa Aesar 40795 graphite powder, -200 mesh, 99.9995%), an investigation of the possible sources of lithium will also be presented. The performance of the 1 MV system will be reviewed and compared to other AMS systems.

Topic : NFF 11

Session 6B

## The New HVE 1 MV Multi-Element AMS system installed at the Aarhus AMS Dating Centre.

Heinemeier Jan,<sup>1</sup> Olsen Jesper,<sup>1</sup> Klein Matthias,<sup>2</sup> Mous Dirk.<sup>2</sup>

[1]AMS  $^{14}\text{C}$  Dating Centre Aarhus, (Denmark)

[2]High Voltage Engineering Europa, (Netherlands)

Aarhus University, Department of Physics and Astronomy has installed a new multi-element AMS system at its AMS  $^{14}\text{C}$  Dating Centre. It is manufactured by High Voltage Engineering Europa B.V. and based on a 1 MV Tandetron accelerator with a dual gas system (Ar and He) for the terminal stripper to investigate possible increased charge exchange efficiency for e.g. actinides. The injector is equipped with two independently operating ion sources and a 120o bouncer magnet with high resolution and a bending power of 340 amu at 35 keV, supporting the measurements of actinides. The high-energy (HE) spectrometer features a degrader foil for isobar suppression and a second HE magnet for suppression of ions scattered in the HE ESA. The control system supports different methods for isotope switching : "traditional" fast bouncing, adjusting the Hall-probe controlled magnet fields (for e.g.  $^3\text{H}$ ) or changing the complete set of operation parameters (e.g. for actinides). During the on-site acceptance tests, the following background levels were measured :  $< 10^{-16}$  for tritium,  $< 10^{-15}$  for  $^{14}\text{C}$ ,  $< 10^{-15}$  for  $^{10}\text{Be}$  (down to  $< 10^{-16}$  in a later 3 hour run),  $2 \times 10^{-15}$  for  $^{26}\text{Al}$ ,  $2 \times 10^{-13}$  for  $^{129}\text{I}$  and  $9 \times 10^{-12}$  for  $^{41}\text{Ca}$  and for  $^{239}\text{Pu}$  ( $^{240}\text{Pu}$ ) a 1.5 (0.5) pg per mg iron, which demonstrates the multi-element capability of the system. System design and performance parameters will be presented.

Topic : NFF 01

Session 6B

## Performance evaluation of the new DirectAMS geochronological sample preparation laboratory.

Zoppi Ugo,<sup>1</sup> Adler Jonathan,<sup>1</sup> Chatters Jim,<sup>1</sup> Wegeleben Emily,<sup>1</sup> Tate Alyssa,<sup>1</sup> Sullivan Molly.<sup>1</sup>

[1]Accium BioSciences, (United States)

The Accium BioSciences AMS facility has been in operation since 2006 and over 55,000 targets have been measured thus far. In early 2011, Accium decided to extend its biomedical core business to provide AMS services to the broader scientific community. The resulting influx of samples (in 2013 geochronological application accounted for approximately half of the number of measured unknowns) necessitated the build-out of a dedicated sample pre-treatment and graphite production laboratory. This new facility is now fully operational and a wide variety of samples types (including wood, charcoal, sediments, shells and bones) is routinely processed. In this paper we describe its features and present results of a series of quality control samples of known age. Particular emphasis is placed on the characterization of the extraneous carbon introduced during sample preparation.

Topic : NFF 02

Session 6B

**”Laboratorio de Espectrometría de Masas con Aceleradores” (LEMA) : New AMS Facility in Mexico.**

**Chavez Efrain,<sup>1</sup> Solis Corina,<sup>1</sup> Barron-Palos Libertad,<sup>1</sup> Ortiz Maria,<sup>1</sup> Andrade Eduardo,<sup>1</sup> De Lucio Oscar,<sup>1</sup> Huerta Arcadio,<sup>1</sup> Araujo Victoria,<sup>1</sup> Marin Laura.<sup>1</sup>**

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LEMA is a new AMS facility in Mexico with the initial capacity to measure low concentrations of  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and Pu isotopes. In this presentation, the main features of the facility are described along with some of the first applications in fields like Archeology, Geology and Nuclear Astrophysics.

Topic : NFF 04

Session 6B

**The new 6 MV multi-nuclide AMS facility at the University of Tsukuba.**

Sasa Kimikazu,<sup>1</sup> Takahashi Tsutomu,<sup>1</sup> Matsunaka Tetsuya,<sup>1</sup> Matsumura Masumi,<sup>1</sup> Satou Yukihiro,<sup>1</sup>  
Izumi Daiki,<sup>1</sup> Sueki Keisuke.<sup>1</sup>

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A new AMS facility has been designed and constructed at the University of Tsukuba after the Great East Japan Earthquake. The AMS system consists of the 6 MV Pelletron tandem accelerator, two 40 multiple cathode AMS ion sources (MC-SNICS), a dedicated AMS beam line and a rare particle detection system. The 6 MV Pelletron tandem accelerator will also be applied to nanotechnology science, IBA, heavy ion irradiation, nuclear physics and so forth. We introduce a CO<sub>2</sub> gas introduction system for one of the two MC-SNICS ion sources for <sup>14</sup>C AMS. The magnets at the high energy side have a mass energy product of  $ME/Z^2 = 176$  amu MeV. A 22.5° electrostatic cylindrical analyzer (ECA) with a 3.8 m radius is provided to filter out unwanted ions. The ECA has a resolution of  $E/\Delta E = 200$ . The rare particle detection system with a five-electrode gas detector is provided to measure the rate of energy loss in the gas for each particle entering the gas detector chamber. The rare particle detection system will be capable of measuring environmental levels of long-lived radionuclides of <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca and <sup>129</sup>I. The 6 MV AMS system is also expected to measure other radionuclides such as <sup>32</sup>Si, <sup>90</sup>Sr, and so on. The 6 MV AMS system was installed in the spring of 2014 at the University of Tsukuba. Beam delivery will start in September 2014. In this presentation, the performance of the 6 MV AMS system and the research programs at the University of Tsukuba will be reported.

**The new AMS Centre for Accelerator Science at ANSTO - a vision to the future.**

**Fink David,<sup>1</sup> Hotchkis Michael,<sup>1</sup> Wilcken Klaus,<sup>1</sup> Child David,<sup>1</sup> Fujioka Toshiyuki,<sup>1</sup> Jacobsen Geraldine,<sup>1</sup> Levchenko Vladimir,<sup>1</sup> Mifsud Charles,<sup>1</sup> Smith Andrew,<sup>1</sup> Williams Alan.<sup>1</sup>**

[1] Institute of Environmental Research, Sydney, (Australia)

In 2009, the Australian Federal Government approved funding for the construction of a state-of-the-art AMS and IBA centre for applied accelerator science at ANSTO. The final design includes a 1 MV dedicated AMS system ( $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{10}\text{Be}$ ,  $^{129}\text{I}$ , Pu, U), a 6MV shared IBA and AMS system ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{129}\text{I}$ ), a gas-filled-magnet beam line, and a dedicated AMS sample preparation building. The NEC accelerators are housed in a new 1500 m<sup>2</sup> building interconnected with the existing ANTARES AMS Facility. The 1200 m<sup>2</sup> chemistry building was designed as a multi-purpose laboratory which provides dedicated and separated lab space for sample preparation for cosmogenic  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$ , meteoric  $^{10}\text{Be}$ , low-level actinides (Pu,U) and an ice-core freezer storage facility for  $^{14}\text{CH}_4$  and  $^{10}\text{Be}$  work. An independent Uranium series laboratory (for carbonates, water, quartz) is available with allocated labs for an ICP-MC and IRMS units. The 1MV AMS accelerator was delivered in October 2013. Large beam-optical acceptance and high-mass resolution analysers, coupled to a novel fast coupled HE and LE isotope switching system, enables high quality radiocarbon and actinide analyses. The 6MV AMS System has 3 ion-sources (alphatross, duoplasmatron, MC-SNICS), a high-mass resolution injector, and 3 AMS beam lines. Construction of all facilities has been completed, and delivery of the NEC 6MV accelerator is scheduled for mid-2014. The 1 MV AMS is operational and first results are presented in a companion conference paper. The gas-filled magnet beam line has been assembled and tested with stable beams. The AMS sample chemistry building is operational and commissioned. The cosmogenic lab covers an area of 250 m<sup>2</sup>, and allows parallel sample processing.



Topic : SP 06

Session 6B

**Development of ultrasensitive  $^{10}\text{Be}$  analysis at MALT.****Horiuchi Kazuho,<sup>1</sup> Matsuzaki Hiroyuki.<sup>2</sup>**

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[2]MALT, The University of Tokyo, (Japan)

Cosmogenic  $^{10}\text{Be}$  serves as a chronometer of rocks of the earth, a proxy of the past cosmic-ray variations, a tool for synchronization among a variety of paleoenvironmental archives, and tracers for various climato-sedimentary processes. In almost all of these cases, a significant decrease of sample quantity indispensable for the analysis has great benefits : it enables us to do ultra-high resolution studies, expands measurable samples into those having an ultra-trace  $^{10}\text{Be}$  concentration, and facilitates analyses of very precious materials. We had already demonstrated an ultrasensitive  $^{10}\text{Be}$  analysis with the Accelerator Mass Spectrometry (AMS) system at MALT, The University of Tokyo, where measurement using 1 mg of hemipelagic sediments and 30  $\mu\text{g}$  of  $^9\text{Be}$  carrier was achieved (Horiuchi et al. 2012). For  $^{10}\text{Be}$  analyses with AMS,  $^9\text{Be}$  carrier is spiked into the samples in a pretreatment stage and the ratios of  $^{10}\text{Be}$  in samples and the spiked  $^9\text{Be}$  are measured with an AMS machine. Therefore, the achievement of the ultra-high sensitivity  $^{10}\text{Be}$  analysis depends on an optimization (a minimalization) of the carrier (i.e. How we can properly decrease the quantity of the  $^9\text{Be}$  carrier indispensable for AMS) and the total efficiency in the AMS system, as well as a use of a low background carrier. In this study, we performed a set of experiments to explore the possibility of further reducing weight of the  $^9\text{Be}$  carrier (10  $\mu\text{g}$  or less). Main points of our examinations are as follows : 1) What is the optimized ratio of Be and Nb for making the AMS target for the ultrasensitive analysis ? 2) What is the suitable strategy for the AMS operation and data analysis ? 3) Other improvements on the sample pretreatment. Those all will be discussed in this presentation.

**17.4 Tuesday 26 August - Afternoon****Topic : AAT 10****Poster Session 1****Isotopic Enrichment of AMS Sample by Using Electrochemistry Technique.****Ouyang Yinggen,<sup>1</sup> Jiang,<sup>1</sup> He Ming,<sup>1</sup> Dou Liang,<sup>1</sup> Xiaoming Wang,<sup>1</sup> Kejun Dong.<sup>1</sup>**

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Accelerator Mass Spectrometry (AMS) has been one of the most promising methods for measuring long-lived radionuclides. However, the detection sensitivity is still hard to satisfy the measurement of some nuclides at natural level due to the extremely low concentration such as superheavy element (SHE) and so on. A special electrochemical method was recently developed via enriching of ekalead elements at the China Institute of Atomic Energy (CIAE). Two projects are carried out, the first is in searching for SHE in terrestrial materials by means of AMS, second one is that enrichment of  $^{41}\text{Ca}$  isotopic abundance its dating and archaeology applications. The principle, experimental setup, performances and main results of the first electrochemical method application in AMS measurement will be detailed in this contribution.

Topic : AAT 11

Poster Session 1

## Facing Social Demands to Develop AMS Techniques and Set up Methodologies for Application.

Jiang Shan,<sup>1</sup> He Ming,<sup>1</sup> Dong Kejun,<sup>1</sup> Dou Liang,<sup>1</sup> Wang Xiaoming,<sup>1</sup> Yang Xuran,<sup>1</sup> Hu Yueming,<sup>1</sup> Bao Yiwen,<sup>1</sup> You Qubo,<sup>1</sup> Ouyang Yinggen,<sup>1</sup> Wang Shilian,<sup>2</sup> Li Qi,<sup>2</sup> Liu Guangshan,<sup>3</sup> Xie Linbo,<sup>3</sup> Shen Hongtao,<sup>4</sup> Pang Fangfang,<sup>4</sup> Ruan Xiangd,<sup>5</sup> Yongjing Guan,<sup>5</sup> Jiliang Wu,<sup>6</sup> Xinchao Lin,<sup>7</sup> Jianhui Qin.<sup>7</sup>

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It has become a very important tool for analysis of long-lived nuclides science 1980's AMS was founded. Especially  $^{14}\text{C}$  has made a very big contribution for its AMS measurement and applications. At the present and future, what is the mission for our AMS scientists? It will be to facing and to meeting the social demands which should be not only to develop AMS techniques but also to set up its application methodologies. The social demands include energy and its safety, human healthy, environment and climate change, resources and also many fundamental sciences and so on. In order to meet the demands, in one hand the AMS techniques need to be developed which will be to increase sensitivity, miniaturization and high precision, fast and on line measurement so on. On the other hand, we have to make efforts for methodologies study for technique application. The social demands in detail will be discussed and the development both at AMS techniques and its application methodologies at the China Institute of Atomic Energy will also be introduced. The developed techniques include, a small AMS system for heavy nuclides measurements, the fast sample preparation and measurement, to combination of AMS and other sampling techniques such as electrochemistry, and so on. The methodologies for AMS applications were also set up and developed under the collaboration with related filed of scientists which include, fast and high sensitivity measurement in nuclear energy and nuclear safety by using  $^{129}\text{I}$ ,  $^{14}\text{C}$  and  $^{135}\text{Cs}$ , tracing for disease diagnose and treatments in human and fruits by  $^{14}\text{C}$  and  $^{41}\text{Ca}$ , tracing for resources of underground water and petroleum with  $^{14}\text{C}$ , dating of Fe-Mn crust in ocean with  $^{129}\text{I}$  dating and archaeology with  $^{41}\text{Ca}$  and so on.

Topic : AAT 12

Poster Session 1

**A Large-acceptance Tandetron AMS System (Part 2).****Von Reden Karl,<sup>1</sup> Longworth Brett,<sup>1</sup> Long Patricia,<sup>1</sup> Roberts Mark.<sup>1</sup>**

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At the 21st Radiocarbon Conference in Paris we presented the first stage of a major upgrade of the National Ocean Sciences AMS Facility (NOSAMS) Tandetron AMS system. It consisted of the replacement of the simultaneous (Recombinator) injector with a fast-cycling sequential injector and changes to the low-energy acceleration section. First data presented in 2012 already showed a large improvement in the system's background data. Over a year of operation with the new injector has confirmed this finding : We now consistently analyze  $^{14}\text{C}$ -free commercial graphite at the low to mid  $10^{-16}$  level for the  $^{14}\text{C}/^{12}\text{C}$  ratio, down almost an order of magnitude from pre-upgrade times. Improvement in the accuracy of the analyses is now also seen in the results for secondary standards. The second stage of the upgrade was delayed due to funding shortfalls : the replacement of the original sputter ion source with an MC-SNICS had to be postponed until now. Apart from operational simplification (the two AMS systems at NOSAMS will have the same ion source technology) the important improvement is the increased beam energy that can be achieved with the new source. That is expected to improve the transmission through the system and reduce the precision-limiting isotope ratio dependence on beam current we now observe for large beam differences. In preparation for the future installation of a 134-sample MC-SNICS (manufacturer lead time 24 months), we have entered into a loan agreement with UC Irvine to transfer their unused 40-sample MC-SNICS to NOSAMS. It is presently being installed and tested. At the time of this meeting we expect to have several months of operational data for this new configuration.

Topic : AAT 23

Poster Session 1

## Simple experiments to modestly improve $^{27}\text{Al}$ currents with the CAMS high-intensity Cs sputter source

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Measurement of  $^{26}\text{Al}/^{27}\text{Al}$  in geological samples has long been recognized as a potentially powerful tool for geochronology, but at present, relatively low beam currents for the stable  $^{27}\text{Al}$  isotope present the major hurdle to its widespread application for cosmogenic exposure and burial dating. At many AMS facilities,  $^{27}\text{Al}$  currents are an order of magnitude or more lower than currents on  $^9\text{Be}$ ; for example, at CAMS over the last two years,  $^{27}\text{Al}$  currents have been at most 1-2  $\mu\text{A}$ , compared to 20-30  $\mu\text{A}$  for  $^9\text{Be}$  (both mixed with Nb powder). This results in much less precise measurements and long counting times, preventing high-precision calculations of young exposure ages or old burial ages, and limiting throughput of samples. While a number of methods are being tested for improving  $^{27}\text{Al}$  currents by an order of magnitude or more, these generally represent significant investments (i.e. gas-filled magnet or Al- metal preparation; e.g. Fifield et al. 2007, NIM-B, 259 :178-183). However, even a relatively modest 2- to 4-fold increase in currents would improve the situation. Initial testing of cathode packing depth and observations during recent measurement campaigns indicate that samples packed somewhat fuller produce higher currents (Hunt et al. 2007, NIM-B, 260 :633-636), but the influences of depth in the cathode hole and Cs focusing distance in the ion source have not been separated. Here we present the results of experiments varying the mixing proportion of  $\text{Al}_2\text{O}_3$  to Nb, the packing depth of the mixture in the cathode hole, the Cs focusing distance, and cathode loading technique (e.g. pounding vs. pressing) with the goal of making modest, but significant, increases in  $^{27}\text{Al}$  currents.

Topic : AAT 24

Poster Session 1

## The Use of AlN to Improve $^{26}\text{Al}$ Accelerator Mass Spectrometry Measurements and Production of Radioactive Ion Beams

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We present results and discuss the use of AlN as an optimal source material for AMS and Radioactive Ion Beams (RIB) science applications of  $^{26}\text{Al}$  isotopes. The measurement of  $^{26}\text{Al}$  in samples by accelerator mass spectrometry is typically conducted on  $\text{Al}_2\text{O}_3$  targets. However,  $\text{Al}_2\text{O}_3$  is not an ideal source material because it does not form a prolific beam of Al required for measuring low-levels of  $^{26}\text{Al}$ . Multiple samples of  $\text{Al}_2\text{O}_3$ , AlN, mixed  $\text{Al}_2\text{O}_3$ -AlN as well as  $\text{AlF}_3$  were tested and compared using the test stand and the stable ion beam (SIB) injector platform at the 25-MV Tandem at ORNL. Negative ion currents of atomic and molecular aluminum were examined for each source material. It was found that pure AlN target produced substantially higher beam currents than the other materials and that there was some dependence on the exposure of AlN to air. The applicability of using AlN as a source material for geological samples was explored by preparing quartz samples as  $\text{Al}_2\text{O}_3$  and converting them to AlN using a carbo-thermal reduction technique, which involves reducing the  $\text{Al}_2\text{O}_3$  with graphite powder at  $1600^\circ\text{C}$  within a nitrogen atmosphere. The quartz material was successfully converted to AlN. Thus far, AlN proves to be a promising source material and could lead towards increasing the sensitivity of low-level  $^{26}\text{Al}$  AMS measurements. The potential of using AlN as a source material for is also very promising to produce more intense radioactive beams of  $^{26}\text{Al}$ .

\*Research sponsored by the LDRD Program at ORNL

Topic : AAT 25

Poster Session 1

**160 keV  $^{26}\text{Al}$ -AMS with a single-stage accelerator mass spectrometer****Shanks Richard,<sup>1</sup> Freeman Stewart.<sup>1</sup>**

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Proof-of-principle  $^{26}\text{Al}$ -AMS sufficient for environmental sample analysis is achieved with a single-stage accelerator mass spectrometer (SSAMS) utilising very low ion energy. The SSAMS operates by discriminating against atomic isobar interference in a negative ion source and suppressing molecules with thick gas stripper. Resulting  $1^+$  ions counting is with a surface barrier detector. The NEC SSAMS for  $^{14}\text{C}$  analysis is a popular model accelerator mass spectrometer and the developed further capability might be a significant addition to established  $^{26}\text{Al}$ -AMS capacity.

Topic : AAT 26

Poster Session 1

**Electrostatic deflector system for digital AMS fast pulsing****Palonen Vesa,<sup>1</sup> Tikkanen Pertti.<sup>1</sup>**

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We have developed and installed electrostatic deflectors before and after the injection magnet to enable the fast pulsing of isotopes. The sequential pulsing of the isotopes, stable isotope current measurements, and rare isotope detection are all performed with three real-time NI-PXI computers. The three computers share time, i.e. their clocks synchronized to better than 100ns, making it easy to commence the data acquisition at different parts of the system when the corresponding isotope is put through the accelerator. Currently we rotate between the three carbon isotopes with a roughly 70Hz cycle and attain precision of 0.3% for most samples.



Topic : AAT 27

Poster Session 1

## A large phase-space acceptance radio-frequency quadrupole ion guide and deceleration system for AMS ion beams

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Over the last two decades several groups using accelerators for applications such as radioactive ion beam analysis (RIB) and accelerator mass spectrometry (AMS) have incorporated prototype radio-frequency quadrupole (RFQ) instruments into their low energy beam lines for ion-gas cooling and/or isobar suppression through ion-gas interactions. Typically, negative ions are produced in a sputter ion source and accelerated to between several and tens of keV kinetic energy, are analyzed by at least one element such as a magnet, and then injected into the RFQ system before injection into the accelerator. In the RFQ system, ions are generally decelerated to  $< 100$  eV, introduced into an RFQ segment with several mTorr gas, and then re-accelerated for injection into the accelerator. An RFQ system design that achieves large transmission of the analyte ions has proven very challenging. Deceleration optics must accommodate large ion beam phase spaces and kinetic energy spreads. Under appropriate initial conditions, ions can gain much kinetic energy due to the RFQ field and be lost, for example, to scattering or electron detachment. Presented are a design and simulations using the modelling software SIMION 8.1 for a novel system with large transmission for a modern AMS ion beam. Initial ion beam conditions were : diameter 3 mm,  $> 35$  mrad spread, and kinetic energy 35 keV with a Gaussian kinetic energy distribution with full width half-maximum value of 8 eV. Under vacuum conditions, 100% transmission was achieved. A code was written to simulate ion-gas collisions. Ions simulated to have the properties of  $^{36}\text{Cl}^-$  had  $> 97\%$  transmission in 5 mTorr He (centre of mass electron detachment threshold energy of 7 eV) while being cooled to  $< 2$  eV in  $< 400$  mm.

Topic : AAT 28

Poster Session 1

## Technological development for Strontium-90 determination using AMS

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Shibayama Nao,<sup>1</sup> Izumi Daiki,<sup>1</sup> Kinoshita Norikazu,<sup>2</sup> Matsuzaki Hiroyuki.<sup>3</sup>

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Strontium-90 ( $^{90}\text{Sr}$ ) is a fission product and expected to be released into the environment such as a nuclear accident.  $^{90}\text{Sr}$  quantities are usually measured by chemically isolating Sr followed by low-level beta counting, however, there are complicated and time-consuming processes to determine  $^{90}\text{Sr}$  isotope. Rapid and simple processes are highly required to conduct  $^{90}\text{Sr}$  quantities, especially after the nuclear disaster. The Accelerator Mass Spectrometry (AMS) is one of potential methods which would solve these problems. We have attempted to develop the  $^{90}\text{Sr}$ -AMS using the 5 MV tandem accelerator at MALT, the University of Tokyo. In this presentation, we report production tests of negative ion beams and detection tests of  $^{90}\text{Zr}$  with the AMS techniques for effective  $^{90}\text{Sr}$  measurements. Target samples of  $\text{SrF}_2$  were made with chemical separation from ground soil in Japan. Mixed samples of the purified  $\text{SrF}_2$  and  $\text{PbF}_2$ , pressed in an aluminum sample holder, were installed in the MC-SNICS ion source. The negative ions of  $^{88}\text{SrF}_3^-$  with over 500 nA were successfully extracted. Natural isobar of  $^{90}\text{Zr}$  interferes with  $^{90}\text{Sr}$  detection using an ionization chamber. However, in order to observe the behavior of the  $^{90}\text{Sr}$  in gas counter, isobaric of  $^{90}\text{Zr}$  was monitored as  $^{90}\text{Sr}$  in this study. After the ions passed through a gas stripper, the ions with 90 for mass number and  $6^+$  for charge state, which are considered to be  $^{90}\text{Zr}$ , were transported to an ionization chamber. The ions with 1000 cps for count rate were identified in spectra, which correspond to approximately  $3.10^{-8}$  for atom ratio of  $^{90}\text{Zr}/^{88}\text{Sr}$  in the Sr beam. Further works would provide an isobaric separation.

Topic : AAT 29

Poster Session 1

**Progress and development of the PRIME Lab gas-filled magnet system****Woodruff Thomas,<sup>1</sup> Caffee Marc.<sup>1</sup>**

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Isobaric interferences may be reduced with the use of a gas-filled-magnet (GFM) directly in front of a dE/dx detector . The GFM is now in routine use at PRIME Lab.  $^{10}\text{Be}$  measurements are now carried out exclusively on the GFM system. Although the transmission through the GFM is only 70% for  $^{10}\text{Be}$  we have eliminated any boron derived interferences. The suppression of the boron-derived interferences has made our backgrounds reproducibly better. Carrier solutions in our own laboratory and some prepared in collaborators laboratories measure  $\sim 4 \times 10^{-16}$   $^{10}\text{Be}/^9\text{Be}$ . Tests with  $^{26}\text{Al}$ , though promising, have still only produced a maximum GFM transmission of 45%. Although the measured minimum beam size for the isobar  $^{26}\text{Mg}$  in the dispersive plane of the magnet is slightly smaller than anticipated ( 2cm FWHM) we believe that the beam is significantly larger in the non-dispersive direction. We are currently installing a larger detector window in order to increase transmission through the GFM and overall machine efficiency. For these tests we are injecting  $\text{AlO}^-$  with currents substantially higher than we obtain with  $\text{Al}^-$ . Even with the present lower transmission our precision for  $^{26}\text{Al}/^{27}\text{Al}$  measurements will be markedly improved.

Topic : AAT 30

Poster Session 1

**Efficient  $^{41}\text{Ca}$  measurements for biomedical applications**

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$^{41}\text{Ca}$  is an AMS isotope particularly interesting for biomedical applications. We have improved the performance of  $^{41}\text{Ca}$  measurements using low-energy AMS at the 500 kV AMS system TANDY at ETH Zurich for reliable measurements and high sample throughput. The main challenge for AMS measurements of  $^{41}\text{Ca}$  is the interfering stable isobar  $^{41}\text{K}$ . Using  $^{41}\text{CaH}_3^-$  ions  $^{41}\text{K}$  can be completely suppressed in the ion source, however, preparation and handling of  $\text{CaH}_2$  is technically challenging and not suitable for large-scale applications. The use of helium as a stripper gas allows transmissions of above 50% for  $^{41}\text{Ca}^{2+}$  ions. The beam switching system is utilized to correct measured  $^{41}\text{Ca}$  ratios for the remaining  $^{41}\text{K}$  contamination. For this purpose,  $^{41}\text{Ca}^{2+}$  and  $^{39}\text{K}^{2+}$  ions are sequentially injected and counted in the detector. Assuming a natural  $^{41}\text{K}/^{39}\text{K}$  ratio the measured  $^{41}\text{Ca}/^{40}\text{Ca}$  ratios can be corrected for the K content and allows measurements down to  $10^{-12}$  range. We will present here the performance of  $^{41}\text{Ca}$  measurements during a larger project in collaboration with the Institute of Food, Nutrition and Health at ETH Zurich. The objective of this campaign was to study whether adequate vitamin D status may reduce the risk of osteoporosis in older adults. In this study 24 subjects (postmenopausal women) were labeled with  $^{41}\text{Ca}$  and urine samples were collected over a time range of 15 months. 6 month after the start the subjects began to consume vitamin D3 supplements. By the change of  $^{41}\text{Ca}$  concentration in the urine the effects of increasing serum 25 (OH) vitamin D concentrations on calcium transfer rates to bone are studied. In total more than 600 samples were analyzed for  $^{41}\text{Ca}$  which was only possible with reasonable effort using the efficient measurement setup at the TANDY.

Topic : AAT 31

Poster Session 1

**Isotope detection on the ANSTO 1MV AMS system**

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[2]National Electrostatics Corporation (United States)

The new 1MV AMS spectrometer at ANSTO is intended for full mass range AMS, from  $^{10}\text{Be}$  to actinides. To allow measurement of up to 8 isotopes either with the 2-anode ionisation chamber or with off-axis Faraday cups, the high-energy magnet chambers can be biased in synchrony with the LE beam injection sequence. Detector data acquisition is based on waveform digitisation rather than peak sensing digital conversion. For  $^{10}\text{Be}$  work the system includes an insertable foil and subsequent achromat before the ionisation chamber. System characteristics and performance will be presented.

Topic : AAT 32

Poster Session 1

## A new simplified Bragg type gas ionization chamber for AMS applications

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The detection of radionuclides in AMS is commonly performed by solid state detectors (Si PIN diodes and silicon PIPS detectors) or by gas ionization chambers. In general solid state detectors offer a much easier handling compared with gas ionization chambers (GIC), but suffer radiation damage and are limited in resolution especially for heavy projectiles. In a  $\Delta E$ -Eres configuration of the GIC particle identification can be performed. But in many cases a detector only for counting purposes is needed. Based on the experiences made with the recently introduced wire anode GICs for IBA applications [1] a simple and compact prototype of a Bragg type detector without Frisch-grid for low energy AMS systems was built. It consists of a cylindrical housing of 25 mm diameter including the gas supply with a copper plate as anode, which is movable along the detector axis in order to vary the effective detector length. A SiN foil was mounted as entrance window and isobutane as detector gas was used. The ratio between signal amplitude and resolution was optimized and investigated as a function of the detector pressure and bias voltage. First measurements with this prototype showed promising results. An energy resolution for carbon at 1 MeV of 26 keV was achieved, which is only about 25 % higher than state-of-the-art GICs. So, for single ion counting applications the achieved performance would be enough. Therefore a new detector design was elaborated and optimized for the use at the MICADAS system, which is routinely in operation since 2013. The physical processes influencing the signal formation (charge collection) and energy resolution as well as the most recent results will be discussed and presented in this contribution.

[1] Müller et al. NIM B 269 (2011) 3037-3040

Topic : AAT 33

Poster Session 1

**Pulse baseline fluctuation due to isobaric interference in the  $\Delta E$  counter****Akihiro Matsubara,<sup>1</sup> Masayasu Miyake,<sup>2</sup> Akimitsu Nishizawa,<sup>2</sup> Yoko Saito-Kokubu.<sup>1</sup>**

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[2]Pesco Corp. Ltd. (Japan)

A deeper understanding of the energy resolution degradation caused by isobaric interference in a  $\Delta E$  counter will provide a way to further improve isobar suppression. The effect of isobar interference on the pulse trace was investigated with the  $\Delta E$  counter for  $^{10}\text{Be}$ -AMS. To create the  $^{10}\text{B}$  overload condition, the gas pressure,  $P_C$ , of the passive absorber cell located in front of the  $\Delta E$  counter was reduced from 62 Torr to 32 Torr, under  $^{10}\text{Be}^{3+}$  and  $^{10}\text{B}^{3+}$  energy at 16.3 MeV by the 4.8 MV tandem accelerator. It is found that as  $P_C$  is reduced less than 54 Torr, the width of baseline fluctuation,  $\sigma$ , of the  $^{10}\text{Be}$  pulse trace respective to the  $\Delta E$  signal increases several times, broadening the  $\Delta E$  component of the  $^{10}\text{Be}$  peak on the  $\Delta E$ - $E_{Res}$  spectrum (where Res stands for Residual). Behavior of  $\sigma$  as a function of  $P_C$  is very similar to that of the energy loss of the  $^{10}\text{B}$  ion calculated by SRIM. The baseline fluctuation can be attributed to accumulation of the positive charge by the  $^{10}\text{B}$  overload. The remaining positive charge lowers the anode potential through the inefficiency of the Frisch-grid. In fact, the mean  $^{10}\text{Be}$  pulse height under the significant fluctuation is smaller than the height expected by the energy loss of the Be ion evaluated by SRIM. If some instability exists in the charge accumulation and/or loss system, the anode potential can fluctuate around its equilibrium value. Volumetric ion-electron recombination would be a key factor for generating the instability.

Topic : AAT 34

Poster Session 1

## A preliminary study of direct $^{10}\text{Be}^{2+}$ counting in AMS using the super-halogen anion $\text{BeF}_3^-$

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The key of  $^{10}\text{Be}$  measurements by AMS is to effectively suppress the interference of isobar  $^{10}\text{B}$ . In this work, a new method of measuring  $^{10}\text{Be}$  by AMS has been studied. This method uses the super-halogen anion of beryllium,  $\text{BeF}_3^-$ , which inherently suppresses  $^{10}\text{B}$  interference by 4 orders of magnitude because the accompanying  $\text{BeF}_3^-$  anion is produced rarely. This  $^{10}\text{B}$  suppression factor is not as high as the traditional degrader method, but the further  $dE/dx$   $^{10}\text{B}/^{10}\text{Be}$  separation in the final ionization detector was found to result in sufficient total  $^{10}\text{B}$  suppression for  $^{10}\text{Be}^{2+}$  to be counted directly at 6 MeV energies. Although the stripping yield from  $^{10}\text{BeF}_3^-$  to  $^{10}\text{Be}^{2+}$  is not as large as that from  $^{10}\text{BeO}^-$ , this inefficiency is compensated with the degrader foil charge fraction avoided. The efficiency for producing  $\text{BeF}_3^-$  and  $^{10}\text{BeO}^-$  was found comparable during experimentation using  $\text{BeO}+\text{PbF}_2$  mixture samples. The preliminary experiment results of the direct  $^{10}\text{Be}^{2+}$  counting method using the 3MV multi-element system at the Xi'an AMS Center will be presented. Further tests are planned to produce  $\text{BeF}_3^-$  from more suitable sample materials such as  $\text{BaBeF}_4$ , which is very rapid to make and safe to handle for sample preparation.

ACKNOWLEDGMENTS : Project supported by the National Natural Science Foundation of China (Grant No.11205161).



Topic : AAT 35

Poster Session 1

**Absolute  $^{14}\text{C}$  IRMS : 6 Years and Counting****Vogel John,<sup>1</sup> Giacomo Jason,<sup>1</sup> Dueker Stephen.<sup>1</sup>**

[1]Eckert &amp; Ziegler Vitalea Sciences Inc. (United States)

Soon after the installation of a MICADAS AMS at our facility in 2008, we noticed that the measured isotope ratio for  $^{14}\text{C}/^{13}\text{C}$  was 13/14 times the correct value for the NIST OxII  $^{14}\text{C}$  standard (SRM-4990c) and for other well known secondary standards. We presented a theory for this behavior at AMS-12. In short, there are a few distinct energy regions of anion collision with gases or stripper foil for which the anion transmission to cation is linear with energy due to the  $v^2$  dependence of collisional ionization. With the istopes at equal energy, the trasnmissions are then inversely proportional to isotopic mass. We now have 6 years worth of data confirming this absolute measurement of the OxII standard. Even including a period in 2010 when we averaged 2.5% too low in the ratio, 12,000 measurements of 3000 samples over the 6 years average 4% below the absolute ratio. We discuss the one physical modification we made of the instrument and our use of simple calculations from only raw scalar counts to analyze our data. The isotopic independence of output from the ion source that is implied in these measurements is a topic of another presentation. We find no dependence of this ratio on ion current, and we introduce chemical-reaction isobar suppression (CRIS) that halved the background count rate in our simple  $^{14}\text{C}$  detector.

Topic : AAT 36

Poster Session 1

## Development of data analysis and control electronics for the multiisotope beamline at CEDAD

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The installation of the new multiisotope beamline at the Centre for Dating and Diagnostics (CEDAD)-University of Salento, required the development of a dedicated system for the control of the vacuum system, analyzing elements (magnets and ESA) and for the data acquisition from the stable isotopes cups and the dual anode gas ionization chamber. A detailed description of the system is presented both in terms of design features, hardware components and control software. In particular the control software has been completely developed in a Labview environment in order to allow easy tuning of the line and unattended routine operation for the wide range of analyzable isotopes ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and actinides). Automatic routines have been included in order to scan all the elements (magnetic and electric fields, Faraday cups position) and find optimal settings by optimizing the beam transmission through the spectrometer. During routine operations, isotopic ratios are calculated on-line together with the measurement uncertainty (Poisson statistics and data scattering) and stored in a log file for the following off-line analysis.

Topic : AAT 37

Poster Session 1

**DREAMS come true : Dresden SIMS becomes Super-SIMS**

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The DREAMS (DREsden AMS) facility [1,2] has been proven to be very suitable for several kinds of applications [3] based on lighter radionuclides. However, the range of applications shall be broaden by upgrading to a so-called Super-SIMS (SIMS = Secondary Ion Mass Spectrometry). Super-SIMS is a combination of trace element AMS (TEAMS) for the determination of stable elements and isotopes using the spatial-resolution - greater than  $3\text{ }\mu\text{m}$  (x,y) and  $5\text{ nm}$  (z) - of SIMS. Thus, this ultrasensitive analytical method is best-suited for analysing geological samples within our focus of resource technology. To realize the DREAMS Super-SIMS, a commercial SIMS (CAMECA IMS 7f-Auto) is used as an ion source and connected to a 6 MV accelerator of highest energy stability. Additionally, the high-energy setup of DREAMS will be equipped with a time-of-flight detector and an energy detection system. By the complete destruction of molecules detection limits some orders of magnitude better than for traditional dynamic SIMS are expected, i.e.  $10^{-9}$  -  $10^{-12}$  (see e.g. [4]). A dedicated housing around the source guarantees the requirements for stable ion source operation, i.e. stability of temperature ( $< 1^{\circ}\text{C/h}$ ) and humidity ( $< 10\%/h$ ). For reducing vibrations the ion source is installed on a cube shaped block made of gabbro (6.4 t weight ;  $1.4\text{ m} \times 1.8\text{ m} \times 0.86\text{ m}$  size). The whole ion source setup can be set on a negative potential of up to -30 kV to allow for higher transmission.

[1] Akhmadaliev et al., NIMB 294 (2013) 5.

[2] Rugel et al. & Pavetich et al., AMS-13.

[3] Feige et al., Ludwig et al., Ott et al., Rodrigues et al., Smith et al., AMS-13.

[4] Maden, PhD thesis, ETH Zurich 2003.

Topic : AAT 38

Poster Session 1

**Accelerator Mass Spectrometry at Purdue University : Improvements at PRIME Lab**

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The Purdue Rare Isotope Measurement Laboratory (PRIME Lab) is a dedicated AMS research facility measuring  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ , and  $^{129}\text{I}$ . PRIME Lab houses chemical processing facilities for both geoscience and bioscience sample preparation. Recent improvements in the AMS and the associated beam-lines include implementation of a gas-filled-magnet (GFM) detection system, construction of a new injector using electrostatic "bouncing", continued improvement of a new data acquisition system based on the XIA PIXE system, and installation of a new computer control system based on National Instruments cRIO electronics. All  $^{10}\text{Be}$  data is now collected on the GFM system and we anticipate using the GFM for routine  $^{26}\text{Al}$  (injecting  $\text{AlO}$ ) and  $^{36}\text{Cl}$  data collection in the near future. All data acquisition operations now use the XIA PIXE module, a single module that replaces traditional nuclear physics NIM-based modules. To improve the precision of our  $^{10}\text{Be}$  measurements we have done tests using the  $^9\text{Be}^{17}\text{O}$  as a proxy  $^9\text{Be}$ . We have resumed routine  $^{14}\text{C}$  measurements, running the tandem at a terminal voltage of 8 MV, using foil stripping, and selecting the +5 charge state.

This work has been funded primarily by NSF and NASA.

Topic : AAT 39

Poster Session 1

## Charge state distributions and charge exchange cross sections of carbon in helium at 45-260 keV

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For low energy AMS, the conversion from conventional stripper gases like N<sub>2</sub> or Ar to the lighter He was very successful. Not only the stripping yields are increased but also less scattering losses are observed due to the smaller nuclear charge of the target atoms. Although He is more difficult to contain in the stripper housing than heavier gases, the density needed for isobaric molecule destruction can be reached within acceptable vacuum conditions. Earlier experiments at ETH as well as data in the literature suggest a continuously increasing 1+ charge state yield of carbon below 400 keV towards lower energies. This enables the development of more compact radiocarbon facilities using lower accelerating voltages or none at all. To optimize machines for charge state yield versus losses caused by the larger ion beam phase space and increased angular scattering, knowledge of the behavior of charge state yields vs. projectile velocity is crucial. In this study, the transmission of carbon in charge states 1+ up to 3+ was measured in He at around 45 keV and 200 keV with different accelerator systems of ETH Zürich. As one of the main results, it could be shown that the 1+ charge state yield at 45 keV is about 80%. Furthermore, the dependency of the transmission on the gas density was used to calculate charge exchange cross sections which are compared with values from the literature.

Topic : AAT 40

Poster Session 1

**Simulation of ion beam scattering in a gas stripper****Maxeiner Sascha,<sup>1</sup> Suter Martin,<sup>1</sup> Christl Marcus,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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The interaction of an ion beam with an AMS (gas) stripper strongly affects the overall transmission of the system. Elastic scattering with the stripper gas atoms leads to a spatial beam broadening within the gas stripper and lowers the transmission. Energy loss fluctuations cause further spatial beam broadening in the dispersive mass spectrometer elements and therefore also affect mass resolution and beam separation. Furthermore, a bad vacuum in the surrounding beam transport regions (e.g. because of non-optimal stripper tube geometries) can lead to increased machine background due to charge exchange processes in the acceleration section. To better understand all these processes, Monte Carlo simulation programs have been written which cover elastic scattering in the stripper, transport and loss of the ion beam and stripper gas flow out of the stripper tube. This contribution presents the physical concepts behind the simulations and discusses the results. Measured beam profiles and transmission values determined at different ETH AMS facilities are presented and compared to the simulations. The comparison turns out to be in good agreement and shows that the assumptions made and the models used are quite reliable and can therefore be used to optimize existing facilities and design new instruments.

## Radiochemical extraction of actinides in the beach areas near by a mothballed nuclear power plant

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The AMS application in this study was the determination of the actinides content in the beach sediments on the Domitian shoreline, immediately down-drift the Garigliano river mouth, in proximity of the Garigliano Nuclear Power Plant, aimed to characterize this area and identify releases by the decommissioning activities released from the plant. The decommissioning activities can represent a source of different kind of polluted discharges, with toxicological effect for the environment and human health. Many studies have been conducted on off-shore sediments using complex techniques whereas very few reference studies do exist on the upper shore sediments. Therefore an intensive research on identifying the most suitable sites for sampling and the best radiochemical method to apply on this kind of matrix was conducted. The methodologies, appropriate for the sandy matrix, was performed at the Nuthech Laboratory, (Risø Campus, placed in Denmark) where it is used the ICP-MS to detect the  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  isotopes. The method is more elaborate and differs by the addition of a precipitation process, which aims to concentrate the actinides, and then through multiple separations on column, the solution is analyzed through the ICP-MS. It was tested for the AMS analysis for the first time at CIRCE (Centre for Isotopic Research on Cultural and Environmental heritage, placed in Caserta, Italy). The results show that the procedure tested in Italy for the AMS had a good outcome and especially the  $^{240}/^{239}\text{Pu}$  ratio is comparable with those obtained by ICP-MS analysis. The ratios and the concentrations obtained show that the presence of these actinides are the results of a global fallout.

Topic : AHN 13

Poster Session 1

## Radionuclides arising from natural and anthropogenic sources in the Domitia coast, Italy

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An AMS application in this study was the determination of the  $^{14}\text{C}$  of mollusca shells in beach core sediments on the Domitian shoreline, immediately downdrift the Garigliano river mouth, in proximity of the Garigliano Nuclear Power Plant and in the left side of the Volturno river mouth. The aim is to date the sediments in a precise age, whether before or after the Chernobyl accident and to perform an environmental characterization of the radioactive releases of the plant. This informations are given usually from the radiochemical measurements of the  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$  and  $^{236}\text{U}$  isotopes. In the first part of this work we focused to check the presence of  $^{137}\text{Cs}$  and to investigate also the long-lived natural radionuclides as  $^{40}\text{K}$ , the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series decay in the sandy samples, by using gamma spectrometry. Results will be illustrated and discussed. It is possible to hypothesize any contamination of the sediments by the nuclear power plant and these considerations emphasize the importance of multidisciplinary characterization (geological, mineralogical, physical) of the area to better understand the issues present and understand the evolution of the processes that occur in it. To get a comprehensive picture of the health state of the Domitian shoreline, this work was flanked also to another study, about to other kind of nuclear releases (U and Pu) present in the same study area.



Topic : AHN 14

Poster Session 1

## AMS and ICP-MS for actinides and $^{137}\text{Cs}$ results around an Italian Nuclear Power Plant

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Italy built and commissioned 4 nuclear power plants between 1958-1978, which delivered a total of 1500 MW. All four were closed down after the Chernobyl accident following a referendum in 1987. One of the plants was Garigliano, commissioned in 1959. This plant used a 160 MW BWR1 (SEU of 2.3 %) and was operational from 1964 to 1979, when it was switched off for maintenance. It was definitively stopped in 1982, and is presently being decommissioned. We report here details on the chemistry procedure and on the measurements for environmental samples, collected up to 4.5 km from the Nuclear Plant. A comparison between uranium concentration as determined by means of AMS (Accelerator Mass Spectrometry) and by ICPMS (Inductively Coupled Plasma-Mass Spectrometry) techniques respectively at the ANU (Australian National University) and at the Ecovise company in Canberra, Australia, is reported, as well as xPu concentration results. Isotopic ratios by means of AMS for both U and xPu are also reported. Additional contribution from Chernobyl is visible whether are introduced  $^{137}\text{Cs}$  gamma activity measurements.

Topic : AHN 15

Poster Session 1

### Progress in the actinides AMS at CIRCE

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A new actinide line [1], based on a 3-MV AMS pelletron tandem system, is operated at the Center for Isotopic Research on Cultural and Environmental heritage (CIRCE) in Caserta, Italy. In this work we report on the progress made in order to push the uranium abundance and mass sensitivity as down as possible. A terminal voltage scan and the beam emittance were performed in order to determine the best measurement conditions. In order to validate the energy and position determinations of the  $^{236}\text{U}$  ions, the energy calibration of the 16 strip silicon detector was verified by comparing the pulse height defect with the literature values. A good measurement reproducibility is obtained and an uranium background mass of few tens of  $\mu\text{g}$  has been determined. Results on  $^{236}\text{U}/^{238}\text{U}$  isotopic ratio show that the background level of about  $3 \times 10^{-11}$  can be reached using a Time of Flight-Energy (TOF-E) system in conjunction with the 16-strip silicon detector with a flight path of 1.5 m. This value is just slightly better than the upper limit of  $6 \times 10^{-11}$  estimated from the yield distribution vs strip number measured without the TOF-E system. We interpret this result as a consequence of the angular straggling due to the thickness of the carbon foil ( $4 \text{ mg/cm}^2$ ), which deteriorates the spatial separation of the interfering ions with respect to  $^{236}\text{U}$ . Moreover, measurements performed with TOF-E system provided from the ANU [2], with a thinner DLC carbon foil ( $1 \text{ mg/cm}^2$ ) and a silicon surface barrier detector, and a longer path (1.9 m) have been performed. It is now possible to measure  $^{236}\text{U}/^{238}\text{U}$  isotopic ratio with a background level of about  $1 \times 10^{-13}$ .

[1] M. De Cesare et al. Nucl. Inst. and Meth. in Ph. Res. B, 294 (2013) 152

[2] L. K. Fifield et al. Nucl. Inst. and Meth. in Ph. Res. B, 117 (1996) 295

Topic : AHN 16

Poster Session 1

**Developments towards detection of  $^{135}\text{Cs}$  at VERA****Lachner Johannes,<sup>1</sup> Martschini Martin,<sup>1</sup> Priller Alfred,<sup>1</sup> Steier Peter,<sup>1</sup> Golser Robin.<sup>1</sup>**

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Radioisotopes produced in natural or anthropogenic fission are widely used for tracer studies of environmental processes, in nuclear forensics, and are important for nuclear waste disposal. Besides the well-known  $^{137}\text{Cs}$  ( $T_{1/2}=30\text{yr}$ ), the longer-lived sister isotope  $^{135}\text{Cs}$  ( $T_{1/2}=2.3\text{Myr}$ ) is also produced, and the combination of the two isotopes would allow for identifying sources of contamination. Because of its long half-life,  $^{135}\text{Cs}$  cannot be detected via decay counting. The stable isobar  $^{135}\text{Ba}$  presently prevents AMS measurements, but we hope to achieve isobar suppression after the installation of the Ion-Laser-Interaction system (ILIAS) at VERA [Forstner et. al. this conference]. We present a preparatory study on the performance of the VERA AMS facility for  $^{135}\text{Cs}$ . Since the usual caesium sputtering would obscure the  $^{135}\text{Cs}/^{133}\text{Cs}$  ratio of the sample, rubidium sputtering was successfully applied. Partial suppression of  $^{135}\text{Ba}$  is possible with the extraction of  $\text{Cs}^-$ , negative  $\text{Cs}^-$ fluorides [Eliades et al. NIMB 294, 361-363], and  $\text{Cs}^-$ oxides, of which currents of several 10nA were extracted over hours from mg amounts of material. The transmission to high charge states (7+,8+) was tested with argon gas and foil stripping. As expected, no suppression in the multi-anode gas ionization chamber [Martschini et al. this conference] could be achieved, leading presently to a  $^{135}\text{Cs}/^{133}\text{Cs}$  detection limit of ca.  $5 \times 10^{-7}$ .

Topic : AHN 17

Poster Session 1

**Study on the AMS measurement method of  $^{135}\text{Cs}^*$ .**

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The discharge of fission product of nuclear fuel material into environment need rapid and sensitive analysis method especially after the Fukushima nuclear accident. Due to its high fission yield and long half life  $^{135}\text{Cs}$  is an important nuclei in the nuclear environmental study. Accelerator mass spectrometry (AMS) may be the only effective method to measure  $^{135}\text{Cs}$  with high sensitivity. Recently, the measurement method of  $^{135}\text{Cs}$  with AMS was carried out. In order to depress the isobar background, extracting  $\text{CsF}_2^-$  and  $\text{Cs}^-$  from ion source were tested. The results showed that although the beam current of  $\text{CsF}_2^-$  is higher than  $\text{Cs}^-$ , the  $^{135}\text{Cs}$  background of extracting  $\text{CsF}_2^-$  is much higher than  $\text{Cs}^-$ . After that, different chemical forms of Cs were tested to find suitable target material for increasing the beam current of  $\text{Cs}^-$ . According to our preliminary result the sensitivity of  $10^{-11}$  ( $^{135}\text{Cs}/\text{Cs}$ ) can be obtained.

Topic : AHN 18

Poster Session 1

 **$^{129}\text{I}$  towards its lower limits****Vockenhuber Christof,<sup>1</sup> Casacuberta Nuria,<sup>1</sup> Christl Marcus,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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Low-energy AMS is well suited for measurements of the long-lived nuclide  $^{129}\text{I}$  because the interfering stable isobar  $^{129}\text{Xe}$  does not form negative ions, thus high ion energies are not required for discrimination in the final detector. Furthermore, low-energy AMS has the advantage that in combination with helium stripping the most probable charge state can be selected; in our case at the TANDY running at 300 kV we select charge state 2+ with a transmission of > 50%. With a proper spectrometer at the high-energy side interferences of the stable isotope  $^{127}\text{I}$  can be completely eliminated.

Contrary to many AMS nuclides  $^{129}\text{I}$  readily forms negative ions and the overall efficiency is high. The challenges lie more in the ion source where cross contamination can be quite severe due to the volatile nature of iodine. This is particularly of importance when analyzing samples that are influenced from anthropogenic sources because the isotopic ratios can span several orders of magnitude. On the other hand special care must be taken when analyzing samples with low isotopic ratios ( $^{129}\text{I}/^{127}\text{I} < 10^{-13}$ ) or samples with very low iodine content (carrier free samples) due to the very same reason.

The advantages and challenges of low-energy AMS of  $^{129}\text{I}$  with the focus on the issues with cross contamination and its correction will be discussed.

Topic : AHN 19

Poster Session 1

## Time series observations of iodine-129 in seawater at the 100 km south of the Fukushima Daiichi nuclear power plant

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The Fukushima Daiichi nuclear power plant (FNPP) accident was occurred in March, 2011. The large amount of radionuclides was released into the environment. Subsequently, the possibility for the leakage of contaminated water from the FNPP was pointed out. Time series observations of  $^{129}\text{I}$  in seawater have been required to assess the leakage problem as well as to understand the migration of the accident-derived radionuclides in the marine environment. In this study, seawater samples were collected in July 2012, in Feb. and Aug. 2013 and in Feb. 2013 by R/V Seikai at the 70~100km south of the FNPP. Iodine in seawater samples was extracted by solvent extraction technique. Iodine isotopic ratio was measured by AMS at the Aomori Research and Development Center of the Japan Atomic Energy Agency. The salinity of seawater collected in summer (in July 2012 and Aug. 2013) was lower than that in winter (in Feb. 2012 and Feb. 2013). This seasonal variation indicated characteristics of two dominant currents flowing in this study area : Oyashio current in summer and Kuroshio current in winter. The  $^{129}\text{I}$  concentrations in summer were about one order higher than the background level, while those in winter were the same as the background. From these results, we concluded that the contaminated water from the FNPP moved southward with the Oyashio current in summer.

Topic : CNA 04

Poster Session 1

**Distribution of  $^{129}\text{I}$  in surface soil in China.****Fan Yukun,<sup>1,2</sup> Hou Xiaolin<sup>1,2</sup> Zhou Weijian,<sup>1,2</sup> Liang Wangguo.<sup>1,2</sup>**

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With the fast development of nuclear industry in China in the past years, nuclear environment safety is becoming a large concern of the public. As a long half-life (15.7 million years) fission product and the same chemical properties and environmental behaviors with high risk short-lived  $^{131}\text{I}$ ,  $^{129}\text{I}$  can be used as an ideal tool for nuclear safety assessment and tracing the environmental processes. The environmental level of  $^{129}\text{I}$  and its distribution in large area of China have not been reported. Aiming to establish the environmental baseline of  $^{129}\text{I}$  in China, 170 surface soil samples (0-10 cm) collected in 2004 from more than 20 provinces, ranging within  $22^{\circ}$ - $48^{\circ}$  N and covering a large area of Chinese territory were analyzed for  $^{129}\text{I}$  and  $^{127}\text{I}$  concentrations. The measured  $^{129}\text{I}/^{127}\text{I}$  atomic ratios vary between  $2.40 \times 10^{-11}$  and  $5.41 \times 10^{-8}$  and >90% of the samples have values within  $10^{-10} \sim 10^{-8}$ . Compared with those reported in the highly contaminated area (such as the soils in Europe,  $10^{-8} \sim 10^{-6}$ ) and the pre-nuclear level of  $^{129}\text{I}$  ( $1.5 \times 10^{-12}$ ), the measured  $^{129}\text{I}/^{127}\text{I}$  ratios in this study implied relatively mild impact of anthropogenic input in most of Chinese environment. Although the number of samples analyzed in some areas is limited especially in northern China, all these obtained data are able to roughly portray an overall picture of  $^{129}\text{I}$  distribution in China. More work is on the way to fill in the gaps where  $^{129}\text{I}$  is insufficiently investigated.

Topic : CNA 10

Poster Session 1

## Exposure history of sand grains buried in Victoria Valley Dune fields, Dry Valleys, Antarctica

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The McMurdo Dry Valleys, Antarctica, have been ice-free for at least 10 Ma. In Victoria Valley, the largest of the Dry Valleys, permafrosted dune-fields displaying hot desert morphologies occupy an area of  $\sim 8 \text{ km}^2$  with dune thicknesses varying from  $\sim 5$  to 70 meters. High-resolution ground penetrating radar (GPR) imaging of selected dunes reveal numerous unconformities and complex stratigraphy inferring cycles of sand accretion and deflation from westerly katabatic winter winds sourced from the East Antarctic Ice Sheet and anabatic summer winds sourced from the Ross Sea. Samples above permafrost depth were taken for OSL and cosmogenic  $^{26}\text{Al}/^{10}\text{Be}$  burial ages. OSL ages from shallow ( $<1\text{m}$ ) pits range from modern to 1.3ka suggesting that deposition/reworking of the dunes is on-going and their present configuration is a late Holocene feature. The same 7 samples gave a mean  $^{26}\text{Al}/^{10}\text{Be} = 4.53 \pm 5\%$  with an average "apparent continuous"  $^{10}\text{Be}$  surface exposure age of  $525 \pm 25 \text{ ka}$  surprisingly indicating a common pre-history independent of depth. Correcting for minor post-burial production based on OSL ages, the minimum (integrated) burial period for these sand grains is  $0.51 (+0.13;-0.11) \text{ Ma}$  which represents the burial age at the time of arrival at the dune. A possible explanation is that this common burial signal reflects recycling episodes of exposure, deposition, burial and deflation, sufficiently frequent to move all grains towards a common pre-dune deposition history. However, it is unclear over what length of time this processes has been active and fraction of time the sand has been buried. Consequently we are presently analysing a third and stable nuclide,  $^{21}\text{Ne}$ , to determine the total surface exposure period. Further coring below permafrost is planned for austral summer 2015.



Topic : CNA 11

Poster Session 1

## Radiometric and stratigraphic dating using $^{26}\text{Al}/^{10}\text{Be}$ ratio in the Dome Fuji (Antarctica) ice core : a feasibility study

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Meteoric (atmospheric)  $^{26}\text{Al}$  and  $^{10}\text{Be}$  are produced by interactions of cosmic rays with specific elements in the air. Because Ar, the target element of cosmic rays for  $^{26}\text{Al}$ , is a trace gas of the atmosphere (0.93% in dry air), the production rate of  $^{26}\text{Al}$  is only a few thousandths of that of  $^{10}\text{Be}$  (e.g. Raisbeck et al. 1983; Auer et al. 2007, 2009; Horiuchi et al. 2007), which is a product of the two most major atmospheric elements : nitrogen and oxygen. Therefore, it is much more difficult to measure meteoric  $^{26}\text{Al}$  than  $^{10}\text{Be}$  in environmental archives. Nevertheless, a paired use of  $^{26}\text{Al}$  and  $^{10}\text{Be}$  has significant potentials for ice-core sciences. One of these is a radiometric dating of old ices to which stratigraphic or ice-flow-model chronology could not be applied. Because the atmospheric production is similar between  $^{26}\text{Al}$  and  $^{10}\text{Be}$ , an exponential decrease of the  $^{26}\text{Al}/^{10}\text{Be}$  ratio with time, in ordinary cases, represents the difference of the decay constants of the nuclides ( $T_{1/2}$  of the  $^{26}\text{Al}/^{10}\text{Be}$  ratio is 1.45 Myr). Another potential utility of the  $^{26}\text{Al}/^{10}\text{Be}$  ratio is a tracer for detecting astronomic or atmospheric events. Even if such events might disturb the radiometric dating, they might be useful for stratigraphic dating of the ice cores. In either case, it is necessary to understand how the  $^{26}\text{Al}/^{10}\text{Be}$  ratio changed (or was stable) among different climatic stages and in the periods of cosmic-ray events. However, those are unclear yet. In this presentation, we will show the  $^{26}\text{Al}/^{10}\text{Be}$  ratios of certain stratigraphic intervals in the Dome Fuji ice core. Those data not only support a reasonable possibility of future radiometric  $^{26}\text{Al}/^{10}\text{Be}$  dating on the "oldest" ice core but also imply some variations in the ratio that might be useful as stratigraphic time markers.

Topic : CNA 12

Poster Session 1

**Exposure dating surfaces in complexly shielded environments with in situ-produced  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{14}\text{C}$** **Hidy Alan<sup>1,2</sup> Matmon Ari,<sup>2</sup>**

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Exposure dating landforms and deposits with terrestrial in situ cosmogenic nuclides (TCNs) hinges on the ability to determine time-averaged nuclide production rates applicable to the sampled material. For samples acquired from steeply dipping surfaces, or surfaces experiencing complex or extreme shielding, accurately determining production rates can be challenging. To circumvent the computation of shielding and sample geometry corrections, we present a three-nuclide approach using  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{14}\text{C}$ . Using a single quartz sample, exposure ages can be determined independently of both local TCN production rates and surface erosion rate -eliminating two of the typically largest sources of error in exposure age calculations. This method takes advantage of the fixed production ratios of these nuclides within quartz and requires knowledge of inherited nuclide concentrations. Where production rates are more easily quantified, however, it can be inverted to calculate these inheritances. We demonstrate this approach with measurements taken from a near-vertical retreating cliff face and its conjugate boulder pile at Solomon's Pillars, Israel.

Topic : CNA 13

Poster Session 1

**CRONUS-Earth : Cosmogenic nuclide calibration**

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In-situ cosmogenic nuclides have been and continue to be an essential tool for understanding Quaternary landforms. During the last decade, analytical capabilities and measurement precision have improved while the geologic issues being addressed have become more complex. As a result, issues of production rates, reproducibility of results, and laboratory-to-laboratory biases have come to the forefront of recent research. The CRONUS-Earth (Cosmic-Ray prOduced NUclide Systematics on Earth) Project was funded to improve the underlying foundations of cosmogenic nuclide systematics and to provide the cosmogenic user community with a uniform platform for use and interpretation of all cosmogenic nuclides. A consistent interpretation is facilitated by the production of a publicly available calculator, CRONUScalc. Production rates were validated in large part by measurements of samples taken from geologic calibration sites. A variety of scaling models were tested with the data from the calibration sites.

This work was funded by NSF.

Topic : CNA 14

Poster Session 1

 **$^{26}\text{Al}/^{10}\text{Be}$  atom ratio : a possible chronometer for old ice ?****Wild Eva Maria,<sup>1</sup> Kutschera Walter,<sup>1</sup> Michlmayr Leonard,<sup>1</sup> Wagenbach Dietmar,<sup>2</sup> Auer Matthias.<sup>3</sup>**

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The age of old ice in Greenland and the Antarctica is of crucial significance in many questions relevant for climate research, as ice opens a window to the past climate on Earth. The datable time span presently covers the last  $\sim 800$  ka. However, for successful dating a continuous and undisturbed stratigraphy of the ice layers is essential. The probability that this requirement is fulfilled, decreases for ice cores approaching bedrock, where along with the oldest ice, hiatus, folding etc. may show up. Therefore the age of old ice bodies remains essentially unknown unless constrained by a suitable direct dating method. Currently several attempts are made to develop such a direct method. At the VERA laboratory the atmospheric atom ratio of  $^{26}\text{Al}$  and  $^{10}\text{Be}$  has been investigated as a possible chronometer and first results were published by Auer et al. in EPSL (2009). The pre-requisite for the applicability of the  $^{26}\text{Al}/^{10}\text{Be}$  ratio is a closed system behaviour of the ice, i.e. - once deposited on the snow surface - changes in the  $^{26}\text{Al}/^{10}\text{Be}$  ratio ( $T_{1/2\text{eff}} = 1.49$  Ma) should be only due to the difference of the half-lives of both radionuclides. Although test measurements of surface samples were very promising, Auer et al. found unexpectedly high ratios for some deep ice samples from the Antarctic EDML ice core. In a follow up project - besides a considerable improvement of the chemical yield for the  $^{26}\text{Al}$  extraction - further samples from this deep ice core were measured, essentially confirming the previous results of Auer et al. Since chemical fractionation between both nuclides during sample preparation can be excluded, it is likely that a fractionation occurs in the ice. This raises the question whether  $^{26}\text{Al}/^{10}\text{Be}$  determinations offer a suitable chronometer for old ice.

Topic : CRI 07

Poster Session 1

**Preparation of an ASTER in-house  $^{10}\text{Be}/^9\text{Be}$  solution.****Braucher R.,<sup>1</sup> Guillou V.,<sup>1</sup> Bourlès D.,<sup>1</sup> Arnold M.,<sup>1</sup> Aumaitre G.,<sup>1</sup> Keddadouche K.,<sup>1</sup> Nottoli E.<sup>2</sup>**<sup>[1]</sup>Aix-Marseille Université, CNRS-IRD-Collège de France, UM 34 CEREGE, Aix-en-Provence, France (France)<sup>[2]</sup>EDF - CEIDRE (France)

For many years, the commercially available certified National Institute of Standards and Technology standard reference material NIST SRM 4325 is used at ASTER to normalize  $^{10}\text{Be}$  measurements. This standard solution being no longer available, we thus decided to produce an in-house standard from the treatment of naturally  $^{10}\text{Be}$  enriched material to generate a solution whose  $^{10}\text{Be}/^9\text{Be}$  ratio will range between  $10^{-12}$  and  $10^{-11}$ , the accurate and precise value being ultimately calibrated against the remaining NIST SRM4325 reference solution. We first decided to prepare such a solution from 2.5 kg of marine sediment we submitted to an adapted chemical protocol that is commonly used to process  $\sim 1\text{g}$  of sediment. These 2.5 kg were leached 3 days in 12 liters of an HCl (37%) solution which have then been evaporated to 3 liters before any treatments. Afterwards, these 3 liters have been purified using  $1\times 8$  Dowex anion exchange resin and lastly purified by 200 solvent extractions using EDTA, Acetyl Acetone and  $\text{CCl}_4$ . The  $^9\text{Be}$  content of the resulting solution has been determined both by ICP-OES and AMS isotopic dilution. Finally, the solution has been diluted within 500 ml of ICP beryllium standard solution to lead to a normalized to the reference material NIST 4325  $^{10}\text{Be}/^9\text{Be}$  ratio of 0.177 (relative standard error=0.15%, N=37) and to an absolute  $^{10}\text{Be}/^9\text{Be}$  ratio of  $(4.932\pm 0.054) \times 10^{-12}$  (i.e. :  $\pm 1.08\%$ ;  $1\sigma$  uncertainties). Nearly at the end of this long preparation, a  $^{10}\text{Be}$  enriched solution originating from a nuclear power plant was made available to us. This gives us the opportunity to prepare another standard solution in greater quantity. Calibrated against NIST standard reference material, the obtained solution which will become the new ASTER  $^{10}\text{Be}$  normalizing reference material leads to a normalized to the reference material NIST 4325  $^{10}\text{Be}/^9\text{Be}$  ratio of 0.428 (relative standard error=0.14%, N=133) and to an absolute  $^{10}\text{Be}/^9\text{Be}$  ratio of  $(1.193\pm 0.013) \times 10^{-11}$  (i.e. :  $\pm 1.08\%$ ;  $1\sigma$  uncertainties).

Topic : CRI 08

Poster Session 1

## Start on RICE-W (Radiocarbon Intercomparison on Chemical Experiments, Water series) program

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We generally use some sample preparation methods for radiocarbon analysis of dissolved inorganic carbon (DIC) in water samples. One is the precipitation method : water DIC is precipitated into  $\text{SrCO}_3$  or  $\text{BaCO}_3$ , and oxidized by  $\text{H}_3\text{PO}_4$  to extract  $\text{CO}_2$ . Another is the bubbling method : water sample is oxidized by  $\text{H}_3\text{PO}_4$  and  $\text{CO}_2$  gas is extracted through bubbling by  $\text{N}_2$  or He gas, according to the WOCE (World Ocean Circulation Experiment) standard method. The other is headspace-extraction method : water sample is oxidized by  $\text{H}_3\text{PO}_4$  and  $\text{CO}_2$  gas in a headspace in a vial flows out with He carrier gas to cryogenic trap. These sample preparation methods have good and bad points each. Therefore, we have initiated a Radiocarbon Intercomparison on Chemical Experiments, Water series (RICE-W) program in Japan to examine whether the  $\text{CO}_2$  extraction procedures of water DIC introduce carbon isotopic fractionation and carbon contamination that can bias the  $^{14}\text{C}$  results. Eight water samples for four kinds of surface seawater, ground water, hot spring water, and sodium bicarbonate solution have already provided to six AMS laboratories in Japan and are being carried out comparison of  $^{14}\text{C}$  measurements. We show the result of the RICE-W program in this presentation.

Topic : CRI 09

Poster Session 1

**AMS measurement of  $^{14}\text{C}$  concentration in 700-yr-old pine tree from Yeongwol, Korea****Chi-Hwan Kim,<sup>1</sup> Jang Hoon Lee,<sup>1</sup> Jin Kang,<sup>1</sup> Sujin Song,<sup>1</sup> Myoung-Ho Yun,<sup>1</sup> Jong Chan Kim,<sup>2</sup>**

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Recently, deviations of the  $^{14}\text{C}$  ages of the Japanese tree samples from those of IntCal04 were reported. Considering this report and the close geographical distance between Korea and Japan, it is highly necessary to investigate the  $^{14}\text{C}$  ages of the Korean tree samples to construct the precise calibration curve for dating samples from Korea. In this study, we will investigate the  $^{14}\text{C}$  concentration in rings of an old pine tree from Yeongwol (37° 21' N, 128° 11' E), Korea. The tree was estimated to be about 700 years old by experts and Yeongwol County Government listed it as a protected tree. However, the tree has now disappeared from history, because of the strong wind occurred in March 2010. Currently, we are trying to collect tree-ring samples from the tree and extract alpha cellulose from each ring. Their annual  $^{14}\text{C}$  concentrations will be measured by using accelerator mass spectrometry (AMS) at Seoul National University, South Korea. Consequently, the results will be matched to the calibration curve IntCal04 to provide an absolute timescale.

Topic : CRI 10

Poster Session 1

 **$^{129}\text{I}$  analysis of international inter-comparison samples in the Xián AMS Center****Zhou Weijian<sup>1,2</sup> Fan Yukun<sup>1,2</sup> Chen Ning<sup>1,2</sup> Zhang Luyun<sup>1,2</sup> Xing Shan<sup>1,2</sup> Hou Xiaolin<sup>1,2</sup>**

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Analysis of standard reference materials (SRMs) and participating in inter-comparison are major ways for the quality assurance of the analysis and validation of a new analytical method. In the past years, our laboratory participated in some inter-comparison exercises organized by International Atomic Energy Agency (IAEA) aiming to produce new SRMs, including seaweed (IAEA-446) and marine sediments (IAEA-410, IAEA-412). These samples were analyzed using our routine combustion method followed by solvent extraction for separation of iodine from sample matrix, while IAEA-412 was also treated by alkali fusion method followed by solvent extraction (Zhou et al. 2010; Hou et al. 1999). Our measured  $^{129}\text{I}$  activity concentration in 12 aliquots of IAEA-446 seaweed is  $0.130 \pm 0.020$  Bq/kg, showing a good agreement with the reported values by other laboratories (Pham et al. 2013), and the  $^{129}\text{I}$  concentrations in the IAEA-412 by two methods agree with each other very well, indicating that the two chemical procedures used in our lab can provide equal valid results of  $^{129}\text{I}$  in environmental samples. In addition, the re-analysis of  $^{129}\text{I}$  in seawater SRM (IAEA-418, seawater collected in the Mediterranean Sea in 2001) indicates that our analytical results of  $^{129}\text{I}$  in 4 aliquots in 2012 and 2013 were  $(2.24\text{--}2.48)10^8$  atoms/L, which agree well with the certified value of  $(2.3 \pm 0.2)10^8$  atoms/L in 2009, implying a sufficient stability of  $^{129}\text{I}$  in this seawater SRM during storage of 5 years.



Topic : CRI 11

Poster Session 1

**Iodine Standard Materials : Preparation and Inter-Laboratory Comparisons****Jenson Douglas,<sup>1</sup> Vockenhuber Christof,<sup>2</sup> Adamic M L,<sup>1</sup> Olson J E,<sup>1</sup> Watrous M G,<sup>1</sup>**

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The Idaho National Laboratory (INL) is preparing to enter the community of AMS practioners who analyze for  $^{129}\text{I}$  iodine. We expect to take delivery of a 0.5 MV compact accelerator mass spectrometry system, built by NEC, in the early summer of 2014. The primary mission for this instrument is iodine; it is designed to analyze iodine in the +3 charge state [1]. We have prepared some standard materials, starting with elemental Woodward iodine and NIST SRM 3231 [Iodine-129 Isotopic Standard (high level)]  $10^{-6}$  solution. The goal was to make mixtures at the  $5 \times 10^{-10}$ ,  $5 \times 10^{-11}$ ,  $5 \times 10^{-12}$   $^{129}\text{I}/^{127}\text{I}$  ratio levels, and some unaltered Woodward, in the chemical form of silver iodide. The various mixtures were synthesized independently of each other; there were no serial dilutions involved. Aliquots of these four materials have been submitted to five established AMS laboratories where iodine analyses are routinely performed : ETH (Zurich), CNA (Seville), PRIME (Purdue), LLNL (California), and Isotrace (Toronto.) Results from all five of these laboratories have been received; in general they indicate that the desired  $^{129}\text{I}/^{127}\text{I}$  ratios have been achieved. The results of this informal round-robin exercise are discussed. If the installation of the instrument at INL goes well, we may have some very preliminary results from the new system. An integral part of data reporting is the approach to data reduction. Accordingly, there will also be some discussion of various philosophies of data handling in regard to the use of standards to adjust data, and background subtraction.

[1] Alfimov, V. and H.-A. Synal. Nucl Instrum. Methods Phys Res. Sect. B 268 (2010) 769-772.

Topic : CRI 12

Poster Session 1

## Development of a new reference material for isotopic ratio measurements of plutonium with AMS

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The motivation of this work is to establish a new multi-isotopic plutonium standard for isotopic ratio measurements with AMS, since stocks of existing standard solutions are declining. To this end, standard solutions of each of the individual isotopes  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$  and  $^{244}\text{Pu}$  were obtained from JRC IRMM (Joint Research Centre Institute for Reference Materials and Measurements). These reference materials (IRMM-081a, IRMM-083, IRMM-043 and IRMM-042a) were diluted with nitric acid and mixed in the procedure to obtain a standard solution with an isotopic ratio of approximately 1.0 : 1.0 : 1.0 : 0.10 ( $^{239}\text{Pu}$  :  $^{240}\text{Pu}$  :  $^{242}\text{Pu}$  :  $^{244}\text{Pu}$ ). A challenge in the preparation of the standard was the accurate weighing of the reference materials due to electrostatic charging caused by the appreciable alpha-activity of the reference solutions. From the stock solution produced in this way, samples were prepared for measurement of the Pu isotopic ratios by AMS. These samples have been measured in a round robin exercise between the AMS facilities at CologneAMS, at the ANU Canberra and ETH Zurich to verify the isotopic ratios and to demonstrate the reproducibility of measurements. We will report details of the procedure and the results of the round robin exercise.

**Estimating bone background uncertainties at SUERC using Statistical analysis.**

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At the Scottish Universities Environmental Research Centre (SUERC), we operate two AMS instruments capable of making routine carbon measurements; a National Electrostatics Corporation (NEC) 5 MV tandem accelerator mass spectrometer and a 250 kV single-stage accelerator mass spectrometer (SSAMS), both use 134-position MC-SNICS sources for running samples. In a standard carbon wheel there are 7 interglacial wood samples, these are used to calculate the organic carbon background for each individual wheel. Bone is frequently dated in archaeological studies, and especially for very old bones (more than 40,000 years old) it is critical to have an accurate and precise measure of the material specific background value and its associated uncertainty. In the SUERC radiocarbon laboratory we have obtained a mammoth bone to use as a background bone sample. A small number of mammoth bone samples are now routinely measured in each AMS wheel, resulting in the accumulation of a large number of bone results over time (2013), and within wheels. The difficulty now is to try and estimate the bone background and the error associated with it, to be subtracted from each unknown bone sample being prepared in the laboratory. We would like to combine the bone samples results in each wheel with the running mean of all the bone samples run in a given year and also include the organic wood backgrounds in each wheel to calculate the minimum error for each unknown bone sample in the wheel. The statistical analysis of the bone results has made use of a linear mixed effects model to examine the variation, and to apportion the overall variation between and within wheels. In this way, a final bone background value and its uncertainty that reflect measurement variability are estimated.

Topic : CRI 14

Poster Session 1

**SIRI samples at CNA : measurements at 200 kV and 1000 kV.****Santos Javier,<sup>1</sup> Gómez-Martínez Isabel,<sup>1</sup> Agulló Lidia,<sup>1</sup>**

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Sixth International Radiocarbon Intercomparison (SIRI) exercise has taken place during 2013 and early 2014. 13 samples were distributed for AMS and 5 for radiometric laboratories. At CNA we have prepared and measured the samples in the two existing AMS dedicated facilities : SARA, a 1 MV multielemental AMS system from HVEE, and Micadas, a 200 kV radiocarbon dating system designed by ETH-Zurich. Results are presented for both systems.

Topic : FNS 5

Poster Session 1

**Compound specific radiocarbon and stable isotope ratio analysis of caffeine :  
authentication of source material in beverages**

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Radiocarbon and stable isotope analysis has proved to be an extremely useful tool in determining authenticity of source material and process of formation of many natural products. Many of the most popular beverages on the market today include caffeine in their composition. Stable isotope mass spectrometry has been used to decipher some synthetic precursors but with limited success as sources and processes change in manufacturing, potentially changing the stable isotope signature. An unambiguous means of determining if caffeine is derived from a natural source is by its radiocarbon content. We present isotopic results from a number of caffeine containing products such as coffee, tea and various sports drinks and beverages, for their authentication of naturally derived caffeine. Methods are detailed with regard to extraction, gas chromatographic detection and analysis and preparative fraction collection for compound specific radiocarbon analysis using accelerator mass spectrometry.

**Forensic investigations at CIRCE AMS laboratory**

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The collaboration between CIRCE in Caserta (Italy) and the Italian Scientific Police Service in Rome (Italy) highlighted some issues on forensic topics. The need to accurately  $^{14}\text{C}$  date paper documents from legal processes encouraged the first part of this study. Paper industries utilize pulp wood from different tree species.  $^{14}\text{C}$  measurement in paper foils is an average of its constituent tree ages. The lack of knowledge about tree ages introduces a high variability in  $^{14}\text{C}$  dating. This study aims to estimate the growth age of trees employed for the paper production before the radiocarbon dating. The estimation is performed through identification of paper fiber. Several paper foils of known production year were analyzed allowing the kind of wood identification and radiocarbon dated. Thanks to the knowledge of the mean age of different wood type generally used in commerce and to the identification of wood kind by means of fiber analysis, we could obtain a more accurate  $^{14}\text{C}$  date. The second aspect analyzed concerns the turnover of bone collagen. It is known that lipid fraction of bone has a very short turnover time, while the turnover of collagen is longer and variable, depending on the individual age. For modern bones, as it is the case of forensic studies, it is important to get as close as possible to the exact year of individual death, so that lipid extraction analysis is advisable. However, the combination of collagen and lipid  $^{14}\text{C}$  measurement results could give other important information, such as the individual age at the moment of death. In this study, from the combination of collagen and lipid results several bones of different known individual age and death year, we could estimate the turnover time of collagen, depending on the age of dead.

**Actinides studies on hot particles at the 1 MV CNA AMS facility**

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Following nuclear events such as nuclear weapons tests and nuclear accidents, particles containing actinides are released into the environment. The actinides isotopic signatures of these particles can be used to label the source and gained additional information on their long-term environmental impact, for instance. To date, much information has been published on the physical and chemical speciation of the particles, environmental behaviour and composition for the most conventional actinides ( $^{241}\text{Am}$ ,  $^{239,240}\text{Pu}$ ,  $^{234,235,238}\text{U}$ ). However, due to the lack of abundance sensitivity of the conventional techniques, very scarce information has been published on  $^{237}\text{Np}$  and  $^{236}\text{U}$ . In this work, we present the first comprehensive information on  $^{237}\text{Np}$ ,  $^{236}\text{U}$  and, also,  $^{239,240}\text{Pu}$ , in four different scenarios : fragments of the so-called Trinitite, a mineral produced in the detonation of the nuclear weapon Trinity (1945, Alamogordo, New Mexico) ;  $\mu\text{m}$ -size particles from Palomares (1966, Spain) and Thule (1968, Greenland), where the nuclear fuel of two thermonuclear devices was accidentally spread due to accidents during their transportation ; and  $\mu\text{m}$ -size particles from the former Russian nuclear test site Semipalatinsk. Preliminary results point out to  $^{237}\text{Np}/^{239}\text{Pu}$  atom ratios ranging from  $1 \times 10^{-4}$  to  $8 \times 10^{-4}$ , and  $^{236}\text{U}/^{239}\text{Pu}$  from  $1 \times 10^{-3}$  to  $9 \times 10^{-3}$ . The actinides measurements were performed on the 1 MV AMS system at the Centro Nacional de Aceleradores (CAN, Seville, Spain), whose performance for the heaviest masses has been studied in different works. The procedure used to measure those samples by AMS and the environmental implications of the results will be discussed.

Topic : FNS 8

Poster Session 1

**Plutonium isotope measurements from Guangxi, China****Guan Yongjing,<sup>1</sup> Wang Huijuan,<sup>1</sup> Hu Jinjun,<sup>1</sup> Ruan Xiangdong,<sup>1</sup> Pan Shaoming.<sup>2</sup>**

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Plutonium is present in environment due to a variety of nuclear activities. The Beibu Gulf, located at 17°00'~21°45'N, 105°40' ~ 110°10'E and surrounded by China and Vietnam, is a natural semi-closed sea area in the South China Sea. At present, the transport data of plutonium in South China Sea is relatively few, and the data relate to Beibu Gulf is blank. A new nuclear power plant, Fangchenggang Nuclear Power Plant, will be put in service in 2015. In order to know the background level of plutonium and sources in this region, plutonium isotope concentrations and ratios from seawater, freshwater, sediments and soils were measured using ICP-MS spectrometry. The transport route and sources of plutonium in seawater and soil will be discussed.



Topic : FNS 9

Poster Session 1

 **$^{14}\text{C}$  activities in terrestrial plants on South Korea's Nuclear Power Plants****Jang Hoon Lee,<sup>1</sup> Chi-Hwan Kim,<sup>1</sup> Jin Kang,<sup>2</sup> Sujin Song,<sup>1</sup> Myung-Ho Yun,<sup>1</sup> Jong Chan Kim.<sup>3</sup>**

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South Korea has 4 Nuclear Power Plants (NPP). They are at Hanul (6 units, 37° 5'52.37 N 129°22'20.76 E), Wolsong (5 units, 35°42'48.58 N 129°28'32.69 E), Kori (6 units, 35°19'19.57 N 129°17'33.63 E), and at Hanbit (6 units, 35°24'54"N 126°25'26") and the total 23 reactor units exist. Also, 20 units among them are running currently (3 units are under repairs) and the total possible power capacity is about 21,740 MW. Furthermore, 2 new NPPs (Samcheok, Yeongdeok) are planned to be installed soon. As the number of nuclear reactors increases, it is gradually considered that the influence for the environment near NPPs should be evaluated more severely than ever and an appropriate safeguard should be established. It is known that  $^{14}\text{C}$  is released from a NPP during routine operation as gaseous effluents. As the study and data collection for this purpose, we have collected leaves and silver grass samples at several locations in the vicinity of the 4 NPPs. Currently  $^{14}\text{C}$  AMS measurement for the samples from Hanbit NPPs was done in advance. Then those from rest 3 sites are being pretreated to extract  $\alpha$ -cellulose and the whole  $^{14}\text{C}$  results from the 4 NPPs will be discussed.

**Search for interstellar  $^{244}\text{Pu}$  as a probe for recent heavy-element nucleosynthesis.**

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Direct observation and detection of freshly produced elements is essential for understanding nucleosynthesis. Here, we address the fundamental scientific question "How were the heavy elements from iron to uranium made?" It is clear that half of these elements including all actinides were produced in a rapid neutron-capture process (r-process), requiring high neutron densities. Their sites and history, however, still remain unclear. Candidates are supernova (SN) explosions or neutron-star mergers, the latter at least 100 times less frequent than SN. If continuously produced, the Interstellar Medium (ISM) is expected to build up a quasi-steady state of abundances of radionuclides (with half-lives  $\leq 100$  My). The solar system moves through the ISM and collects dust particles that might eventually be incorporated in terrestrial archives. Presence of  $^{244}\text{Pu}$  (81 My half-life) in the ISM can place strong constraints on recent r-process frequency and production yield. We applied AMS at the VERA laboratory for the search of small traces of live interstellar  $^{244}\text{Pu}$  and  $^{247}\text{Cm}$  (15.6 Myr), archived in a 1.8 kg deep-sea manganese crust during the last 25 Myr. We extended previous measurements in deep-sea manganese nodules and sediments with a substantially improved sensitivity for detecting interstellar  $^{244}\text{Pu}$ . We will present data suggesting much lower abundances than expected from continuous production in the Galaxy, that may point to a rarity of actinide r-process nucleosynthesis sites, compatible with neutron-star mergers or a small subset of actinide-producing SN.

Topic : GAA 08

Poster Session 1

# **$^{14}\text{C}$ contents in early and late woods of annual tree rings after Carrington flare occurrence in September 1859**

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Rapid enhancements of atmospheric  $^{14}\text{C}$  concentrations were observed in AD775 and AD994 from Japanese tree rings (Miyake et al. Nature 486,240 (2012) and Nat. Commun. 4, 1748 ( 2013)). The rise time and increase were approximately one year confined by annual tree ring and 12‰ and 10‰ . Moreover, the event in AD775 was confirmed in annual tree rings of a German oak (Usoskin et al. A&A 552, L3 (2013)) and hence the events are considered as a global cosmic rays phenomenon. Although the source of events is still unclear and controversial issue, large solar proton events are a considerable candidate. For the AD775 event, the fluence of solar proton above 30 Mev  $4.5 \times 10^{10} \text{ cm}^{-2}$  was calculated (Usoskin et al. 2013). On the other hand, for past 450 years the Carrington flare event of September 1859 is the largest solar proton event which the proton flux above 30 MeV is estimated with  $1.9 \times 10^{10} \text{ cm}^{-2}$  as the omni- directional fluence (Shea et al. Adv. Space Research 48, 232 (2006)). Therefore, it is important to investigate the response of  $^{14}\text{C}$  contents in annual tree rings to the large solar proton events and hence to check it for the Carrington flare. Using YU-AMS, we have measured the  $^{14}\text{C}$  contents in the early and late woods from 1858 to 1863, respectively, in the cedar tree rings grown up at Tsuruoka in north part of Japan between 1811 and 1999. The  $\Delta^{14}\text{C}$  of early woods was  $-4.7 \pm 0.99\text{‰}$  on average and it is consistent to the Stuiver single year data  $-4.3 \pm 0.73\text{‰}$ . We describe the sequence of  $\Delta^{14}\text{C}$  in early and late woods comparing with the Stuiver data indicating a little bit enhancement in 1861 after Carrington flare in September 1859.

**AMS analyses of different soil fractions from paleosols buried by tephra, Alaska, USA.****Cherkinsky Alexander,<sup>1</sup> Wallace Kristi.<sup>2</sup>**

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AMS radiocarbon ages were determined on three different fractions extracted from buried soils within soil-tephra stratigraphic sequences in south-central Alaska as an experiment to establish best practices for radiocarbon dating of low-organic matter soils common in this high-latitude region. Typical soils in these environments are described as Bwj/Cox soils occasionally containing thin (1-2 mm) organic Aj horizons. Contamination of soils by local wind-blown material is a concern. A coalfield composed of 15,500 km<sup>2</sup> of Cretaceous age coal-bearing formations is exposed in numerous valleys where the buried soils were collected. AMS <sup>14</sup>C ages on the humic acid fraction are consistently younger than ages of both the bulk soil and residue after extraction. The difference in ages for the humic acid fraction relative to ages on bulk soil range from 60-1130 <sup>14</sup>C yr BP. The age estimates on residue after extraction are 180-4110 <sup>14</sup>C yr BP older than the humic acid fraction ages. Based on the proximity of our field sites to coal-bearing rock deposits, we attribute the older ages of both the bulk soils and residue after extraction to contamination by old carbon from coal, possibly introduced by wind. This study supports the use of AMS radiocarbon dating of the humic acid fraction of soils in order to estimate the age of decayed organic material within the soil that presumably marks the age of burial and avoids suspected contamination by old carbon no matter the source.

## Background levels of radionuclides $^{36}\text{Cl}$ and $^{129}\text{I}$ in surface soils at East Japan before Fukushima accident.

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The long-lived radionuclides  $^{36}\text{Cl}$  and  $^{129}\text{I}$  are generated by the nuclear tests or interaction with cosmic rays. They have descended to ground or sea level surface, and they have remained ground surface afterward. We have measured amount of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  by accelerator mass spectrometry (AMS) before nuclear accident at the Fukushima Daiichi nuclear power plant. We have collected surface soil samples from the Sea of Japan to the Pacific Ocean at the equal-latitude cross-sectional areas ( $37^{\circ}20' \text{ N} - 37^{\circ}30' \text{ N}$ ) in the East Japan. Inorganic chlorine in soil developed an improved leaching process that uses diluted  $\text{HNO}_3$  as an extractant. After leaching from soils, the  $\text{AgCl}$  samples for AMS-target made from the obtained solutions at ordinary treatment. Isotopic ratios of  $^{36}\text{Cl}/\text{Cl}$  were determined by AMS at Tandem Accelerator Complex, University of Tsukuba. Preparation of  $^{129}\text{I}$  target was following ordinary method. Isotopic ratios of  $^{129}\text{I}/\text{I}$  were determined by AMS at MALT, the University of Tokyo. We obtained the distributions of radionuclides  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in surface soils. The measured  $^{36}\text{Cl}/\text{Cl}$  ratios of 34 surface soil samples which were about 0-10 cm in depth from 6 sites at the equal-latitude cross-sectional areas were between  $0.1 \times 10^{-13}$  and  $4.1 \times 10^{-13}$ . It was shown that the  $^{36}\text{Cl}/\text{Cl}$  ratios are lower at both sea sides. The concentrations of  $^{129}\text{I}$  and  $^{129}\text{I}/\text{I}$  ratios in surface soil (0-10 cm) at 28 points were determined to be 0.18 - 1.13 mBq/kg and  $4.3 \times 10^{-9}$  -  $11.7 \times 10^{-9}$ , respectively.

Topic : GAA 17

Poster Session 1

## AMS contribution for measurement of Long Life Radionuclide at very low level in nuclear wastes.

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Waste management is a key issue for nuclear industry. In France, ANDRA (the national agency for nuclear waste management) is in charge to size, build and operate waste repositories. In order to assure the security of these facilities, a list of radio-isotopes and their associated activities has been defined as acceptance criteria by ANDRA to receive them. From this list, some are readily and easily measured while many such as pure beta emitters are difficult to measure, requiring performing long radiochemical process. Due to its high mobility and long half-life, the acceptance limit for  $^{36}\text{Cl}$  is as low as 5 Bq/g, making it as one of the radio-isotope that will define the final repository. In some case, result of the measurement carried out with classical detection technique as Liquid Scintillation Counting (LSC) is under the detection limit, of typically 1Bq/g. In the lack of further data, the detection limit is automatically taken into account to overvalue the waste activity. Over-estimating waste activity could dramatically increase the cost and will penalize the radioactive capacity of the storage facilities. To overcome this problem, the Operator Support Analyses Laboratory has developed analytical procedures to prepare samples to measure the radio isotope of interest ( $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ) by Accelerator Mass Spectrometry (AMS) as low as 1 mBq / g in nuclear wastes matrixes. We will present the result of the comparison of two procedures used in the laboratory for activated steel samples. A simplified radiochemical method has been implemented in order to analyze  $^{36}\text{Cl}$  with a detection limit around 1Bq/g by LSC. A second procedure has been used to prepare sample for AMS measurement. Radiochemical Blank and homemade standard have also been analyzed.

Topic : GAA 39

Poster session 1

## High resolution record of the $^{14}\text{C}$ spike event between AD 773 and AD 774 in coral in China

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Recently, scientists in Japan reported a spike of  $^{14}\text{C}$  by 12‰ between AD 774 and AD 775. In order to clarify the course of the spike,  $^{14}\text{C}$  content from a 1.2-m fossil *Porites* coral (XDH) drilled from the Xiaodonghai Reef (18°12.46'N, 109°29.93'E) from the northern South China Sea in 1997 and sampled in monthly-resolution and biweekly-resolution was analysed. Results showed that the  $^{14}\text{C}$  content increased by  $\sim 15\%$  during the winter of AD 773, and remain elevated for more than 4 months. Then, it increased and dropped down in the following two months, forming a spike of 45‰ high in late spring. The spike seemed deviate the normal changes of atmospheric  $^{14}\text{C}$  content caused by the radiation intensity variation of solar. Collision of a comet containing higher  $^{14}\text{C}$  content with the earth's atmosphere may result in this spike as the comet was recorded by royal celestial observation in Tang dynasty. However, the reason for the spike was still under debate. The further work will be carried out in the tree ring, coral and varve to see the relation between cosmogenic nuclide ( $^{14}\text{C}$ ,  $^{10}\text{Be}$ ) content with extraterrestrial objects.

## Lowland river responses to intraplate tectonism and climate forcing over the last glacial cycle

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Large-scale folding associated with intraplate tectonism effectively steers lowland rivers, such as Cooper Ck in eastern central Australia. We apply cosmogenic exposure dating ( $^{10}\text{Be}$  and  $^{26}\text{Al}$ ) in bedrock, and luminescence in alluvium, to quantify the erosional and depositional response of Cooper Ck where it incises the rising anticline of the Innamincka Dome. The plucking of bedrock joint-blocks during extreme floods governs incision into the Dome; an incision rate of  $17 \pm 8$  mm/ka is estimated using a numerical model calibrated with  $^{10}\text{Be}$  and  $^{26}\text{Al}$ , involving episodic detachment of 1-3 blocks from the bedrock channel boundary. The last big-flood phase is estimated at maximum 110-120 ka. Upstream of the Innamincka Dome long-term rates of alluvial deposition for the last  $\sim 300$  ka are estimated from 47 luminescence dates. Over the period  $\sim 55$ -75 ka Cooper Ck changed from a bedload-dominant, laterally-active meandering river to a muddy anabranching channel network up to 60 km wide. We propose that this shift was a product of base-level rise linked with the slowly deforming syncline-anticline structure, coupled with a climate-forced reduction in discharge. The uniform valley slope along the rising bedrock and subsiding alluvial system represents i) an adjustment between the relative rates of deformation and ii) the ability of enhanced flows during the Quaternary to incise the rising anticline. Hence, tectonic and climate controls are balanced in the longer term.



## Ancient and recent exposure history of chondrules from two highly primitive meteorites

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Chondrules may have spent several million years as free-floating particles in the solar nebula [1], and if so, been exposed to an early cosmic ray irradiation. The search for "pre-irradiation" in noble gas isotopic signatures has, thus, been actively pursued recently [2-4]. Results for two highly primitive CR3 chondrites (MET00426 & QUE99177) [5] are intriguing : 1) They are among the most unmetamorphosed meteorites, most likely to have retained any pre-irradiation record. 2) Target elements were determined by Instrumental Neutron Activation Analysis on the same material used for noble gas analysis. 3) QUE99177 shows no hint for having been part of an asteroidal regolith. 4) Chondrules show both higher and lower cosmic ray exposure than identically-shielded matrix samples. The shortest cosmic ray exposure determined via stable noble gases is an upper limit to the recent cosmic ray exposure age. Further constraints can be obtained via radionuclides such as  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ , which have been analyzed at DREAMS [6,7]. Despite sample masses of only 1.6-1.8 mg for single chondrules, ratios are as high as  $1\text{-}3\times 10^{-12}$  for  $^{10}\text{Be}/^9\text{Be}$  and  $^{26}\text{Al}/^{27}\text{Al}$ , and  $1\times 10^{-13}$  for  $^{36}\text{Cl}/^{35}\text{Cl}$ , clearly distinguishable from blanks. Preliminary evaluation shows that the radionuclides are not in saturation. However, since the meteorites are finds from Antarctica, one also has to consider decay during terrestrial residence. To better constrain this, AMS of  $^{53}\text{Mn}$  is scheduled for the very near future at ANU.

[1] Cuzzi, Nat. Geosci. 4 (2011) 219.

[2] Eugster et al. MAPS 42 (2007) 1351.

[3] Das & Murty, MAPS 44 (2009) 1797.

[4] Roth et al. MAPS 46 (2011) 989.

[5] Beyersdorf-Kuis et al. 44th LPSC (2013) 1999.

[6] Akhmadaliev et al. NIMB 294 (2013) 5. [7] Rugel et al. AMS-13.

Topic : GAA 44

Poster Session 1

## A comparison of distribution maps of $\Delta^{14}\text{C}$ from 2010 to 2013 year in Korea using Ginkgo tree leaves

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$\Delta^{14}\text{C}$  values of leaves of a deciduous tree are same to those of air within error and are used to map out regional scale fossil fuel ratio in air. We collected a batch of ginkgo (*Ginkgo biloba* Linnaeus, a deciduous tree) leaf samples in July from 2010 to 2013 in Korea to obtain the regional distribution of  $\Delta^{14}\text{C}$ . The  $\Delta^{14}\text{C}$  values of the samples were measured using Accelerator Mass Spectrometry (AMS) in KIGAM, Korea. Averages of  $\Delta^{14}\text{C}$  values of Korea decrease about 6‰ annually but decrease rate in between 2010 and 2011 is smaller than others. Further study need whether this is from effect of Fukushima power plant accident in March 11, 2011 or not. The distribution maps of  $\Delta^{14}\text{C}$  were made using measurements of a batch of ginkgo leaf samples in July from 2010 to 2013 in Korea and shows that  $\Delta^{14}\text{C}$  values in western part of Korea is lower than those of eastern part of Korea. This is from that a lot of industrial complex and population are in western part of Korea and westerly's wind from China containing fossil fuel  $\text{CO}_2$  flows in Korean. The maps also show that much low  $\Delta^{14}\text{C}$  values appear in metro cities (Seoul, Dejeon, Daegue, Busan). Lowest  $\Delta^{14}\text{C}$  values is  $-134 \pm 3\text{‰}$  at Sajik tunnel in Seoul and fossil fuel ratio in air of the place is 12.8 %.

## Quantifying rates of soil organic carbon accumulation in black spruce and tundra ecosystems in Alaska using radiocarbon

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The high-latitude regions, where a serious warming is expected, currently store large amounts of soil organic carbon (SOC) in active-layer soils and permafrost, accounting for nearly half of the global below ground OC pool. Despite the importance of these regions in the present C cycle, the soil C fluxes and budget are still only poorly known. Here, we quantify soil C stock and evaluated the C input (I), decomposition rate (k, inverse of turnover time (TT)) and net C accumulation (CA), in tundra and boreal soils using  $^{14}\text{C}$  approaches.

Cumulative SOC stocks in boreal forest are 5.3 and 19.2  $\text{kgCm}^{-2}$ , in surface organic layer (SL, 0-25 cm), and deep organic and mineral layers (DL, 25-70 cm), respectively. Large annual I ( $0.25 \text{ kgCm}^{-2}\text{yr}^{-1}$ ) and relatively slow k (27 years) lead to rapid CA ( $0.05 \text{ kgCm}^{-2}\text{yr}^{-1}$ ) in SL. DL including near-surface permafrost show slower I ( $0.03 \text{ kgCm}^{-2}\text{yr}^{-1}$ ) and TT (617 years) and CA about 20 times slower ( $0.003 \text{ kgCm}^{-2}\text{yr}^{-1}$ ) than SL. Decomposition of SOC (Rh), which in accord with C losses from both SL and DL, was  $0.23 \text{ kgC m}^{-2} \text{ yr}^{-1}$ . This value agreed well with Rh simulated by process-based model. In contrast, large amount of SOC ( $36.4 \text{ kgm}^{-2}$ ) have accumulated over millennia (TT : 4540 yrs) below the thin organic layer in tundra. The CA is close to zero ( $0.003 \text{ kgCm}^{-2}\text{yr}^{-1}$ ), and Rh is  $0.008 \text{ kgCm}^{-2}\text{yr}^{-1}$ . Our results show that the most SOC in tundra was mode of stabilizing OC by permafrost and steady-state SOC stocks under current C balance.

**Annual German oak and bristlecone pine  $^{14}\text{C}$  data from 2796 BP to 2575 BP**

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The radiocarbon calibration curve shows several distinct features in the interval from 2800 BP to 2400 BP : A relatively steep increase in atmospheric  $^{14}\text{C}$  from 2800 to 2650, and a pronounced and sharp dip around 2625 BP, followed by a decrease until 2400 BP. Here we present a series of high precision annual radiocarbon dates obtained from German oak tree rings spanning an interval of 180 years from 2796 to 2575 BP. In addition, a part of the same interval has been re-measured with annual Bristlecone pine wood from the White Mountains, California. Both the Bristlecone pine and the German oak results confirm a very rapid transient decrease in the atmospheric  $^{14}\text{C}$  content over a period of about two decades around 2625 BP. The high-resolution data also reveals more fine structure and relatively strong short-term variation in the calibration curve. Here we present and discuss detailed results from our new annual data and analyze the investigated interval for 11/22 year solar cycles.

Topic : GAA 47

Poster Session 1

## Age determination of large trees with false inner cavities : AMS radiocarbon dating of the Lebombo Eco trail baobab

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Several anomalies were observed in the AMS radiocarbon dating of wood samples collected from inner cavities of large live African baobabs (*Adansonia digitata* L.). Normally, the age values of samples collected from large central cavities should decrease continuously from the cavity walls toward the outer part of the trunk/stem. However, we found that in many cases the age values increase from the cavity walls up to a certain distance in the wood, after which they decrease toward the outer part. The only explanation for these anomalies is that such cavities are, in fact, only natural empty spaces between fused stems disposed in a ring-shaped structure. We named them false cavities. The first African baobab for which we noted these anomalies, that made possible the identification of false cavities, was the Lebombo Eco trail baobab, located in the Limpopo Park, Mozambique. Here we present the complete AMS results of segments originating from 6 long samples from the Lebombo baobab. The dating results indicate that the tree consists of 5 perfectly fused stems that close almost completely a large false inner cavity. The radiocarbon date of the oldest segment was found to be of  $1425 \pm 24$  BP, which corresponds to a calibrated age of  $1335 \pm 15$  years. The dates also indicate that the stems have stopped growing toward the false cavity over the past 550 years.

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Topic : GAA 48

Poster Session 1

## Searching for the oldest Malgasy baobab : AMS radiocarbon investigation of large *Adansonia rubrostipa* and *Adansonia za* trees

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Six of the nine baobab species (*Adansonia* spp.) are endemic to Madagascar. The two species with the largest total population are *Adansonia rubrostipa* Jum. & H. Perrier (Fony baobab) and *Adansonia za* Baill. (Za baobab); each species is represented by well over one million individuals. This research is the very first investigation of the architecture and age of *A. rubrostipa* and *A. za*. Large individuals belonging to the two species, located in Southern Madagascar, were investigated; the primary method of analysis was AMS radiocarbon dating of wood samples collected from their trunks. The results indicate that big specimens of the two baobab species are typically multi-stemmed and exhibit cluster or ring-shaped structures. According to radiocarbon dates, the oldest known baobab of Madagascar is the so-called "Grandmother", a triple-stemmed *A. rubrostipa* individual which grows in the Tsimanampetsotsa National Park. The dating results suggest an age of 1,000-1,100 years. Thus, *A. rubrostipa* becomes the second *Adansonia* species with "millenarian" trees, i.e. trees that can live over 1,000 years.

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## Structure and age of the Grandidier's baobab (*Adansonia grandidieri*) determined by AMS radiocarbon dating

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The genus *Adansonia* belonging to the Bombacoideae, a subfamily of Malvaceae, consists of nine species. Six species are endemic to Madagascar and have a natural distribution only here. The Grandidier's baobab (*Adansonia grandidieri* Baill.) is the biggest and most famous of the six Malagasy baobab species. Grandidier's baobab is classified as endangered species by the IUCN Red List 2006. However, recent high-resolution satellite images demonstrate that the total population of Grandidier's baobab is much larger than previous estimates, surpassing one million individuals. The research presented here is the first investigation of the architecture and age of the Grandidier's baobab. Several very large individuals from the Morombe-Andombiro-Andavadoaka area, including *Tsitakakoike*, the largest Malagasy baobab, were examined; the main method used was AMS radiocarbon dating of wood samples collected from their trunks. The results suggest that, in terms of total wood volume, the largest Grandidier's baobabs might exceed the largest African baobabs (*Adansonia digitata* L.). The performed research also indicates that big Grandidier's baobabs are typically multi-stemmed, with very large and tall closed empty spaces inside their quasi-cylindrical trunks. According to radiocarbon dates, the ages of the oldest Grandidier's baobabs may exceed 1,000 years.

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Topic : GAA 50

Poster Session 1

**Terraces development and  $^{10}\text{Be}$  variability in deeply incised valleys in an arid region****El Saiy A.<sup>1</sup> Aldahan Ala<sup>1,2</sup> Possnert G.<sup>3</sup> Abdelghany O.<sup>1,4</sup> Yi P.<sup>5,6</sup> Chen P.<sup>6</sup>**

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Terraces represent vital economic land resources for agriculture, domestic uses and landscaping in many places in the world. A good example is the terrace systems associated with evolution of Oman Mountains and related landscape in semi-arid region of the northern United Arab Emirates. In this investigation we have selected a characteristic terrace system within a relatively long valley along the north-northeastern United Arab Emirates. These deposits represent economic, cultural and environmental resources in the area with farms and groundwater wells that have been utilized for many hundreds of years. A combination of field geomorphologic mapping coupled with  $^{10}\text{Be}$  determination in different parts of the terrace system was used to interpret the formation of the terraces in terms of sedimentation and tectonic effects in the area. The results indicate two major cycles of sedimentation in the development of the terrace system that reflect different sedimentary particle shapes, sizes and mineralogy. The  $^{10}\text{Be}$  concentrations in samples show a range of  $(0.35\text{-}2.0)\times 10^8$  atoms/g where the highest concentration is found in the youngest terrace beds. Although, it is difficult to construct a straightforward  $^{10}\text{Be}$  chronology, the results suggest that large span of time (likely over a million years) have associated the terrace development in the area.



**Annually resolved  $^{10}\text{Be}$ -solar activity in lake sediments, 1397-1980 AD**

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There is no doubt that  $^{10}\text{Be}$  is a main tool providing information related to the activity of the Sun and cosmic particles and the consequent impact on the Earth's climate in the past. Investigating pathways and causes of  $^{10}\text{Be}$  historic events will improve prediction of rate of climate change in the future. Ice cores provided leading records of paleoclimate, but retrieving annually-resolved sediments opens new possibility for the extraction of solar activity cycles from terrestrial environments. We report here on the extension of previously published  $^{10}\text{Be}$  record in varved sediments by adding 71 years (extending the record from 1468 to 1397 AD). The data was further compared to annual  $^{10}\text{Be}$  variability in Dye-3 and NGRIP ice cores, Greenland. The sediment core is from lake Lehmilampi in eastern Finland (surface area 0.15 km<sup>2</sup>) which is fed by two streams, and has one outlet stream with a catchment area of about 1 km<sup>2</sup>. The sediments are composed of detrital mineral fraction and mainly in situ organic dominated fraction. Chronology of the sediment was established by varve counting with an estimated cumulative counting error of 2% over the last 2000 years. The results indicate  $^{10}\text{Be}$  concentrations of (3.5-8) × 10<sup>8</sup> atoms/g and show a trend of increasing concentration between 1410 AD and 1480 AD in a manner comparable to the ice cores. The  $^{10}\text{Be}$  flux, however, reveals a rather constant trend during the period 1400-1500 AD, but it is still a decreasing trend when compared with the 1800-2000 AD. The relatively high concentration during the period 1410-1480 AD may reflect a climate shift in the catchment area associated with increasing precipitation. Alternatively change into a period of low solar activity is also possible, but it is not observed in the  $^{10}\text{Be}$  flux data.

Topic : GAA 52

Poster Session 1

## Radiocarbon dating of glycerol dibiphytanyl glycerol tetraether lipids (GDGTs) in the western Arctic Ocean sediments

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In the Arctic Ocean, it is not easy to make age model of these cores because carbonate fossil such as planktonic foraminifera is very limited in sediment. Thus, paleoclimate history of the Arctic Ocean environment is poorly understood. Archaeal and/or bacterial glycerol dibiphytanyl glycerol tetraether lipids (GDGTs) found in surface sediments in Chukchi Sea, Arctic Ocean are derived from various sources from both marine and terrestrial sources, which index using some compounds of GDGTs are used for reconstruction of paleo sea surface temperatures (TEX86). We presented radiocarbon ages of GDGTs as well as carbonaceous fossils and bulk organic matter. In the conference, we will discuss sources of GDGTs and potential of chronological tools for paleoclimate study.

Topic : GAA 53

Poster Session 1

**AMS radiocarbon dates for tephra layers in Adak Island, Aleutian Islands, Alaska****Okuno Mitsuru,<sup>1</sup> Wada Keiji,<sup>2</sup> Torii Masayuki,<sup>3</sup> Tohru Danhara,<sup>4</sup> Nakamura Toshio,<sup>5</sup> Saito-Kokubu Yoko,<sup>6</sup> Gualtieri Lyn,<sup>7</sup> Brenn Sarata.<sup>8</sup>**

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Adak Island, a member of the Andreanof Islands in the Aleutian Islands, Alaska is covered with Holocene sequences of soil-tephra complexes. Tephra layers are useful for establishing a chronographic framework on the island. Black (1976) described the three conspicuous tephra deposits (Main, Intermediate and Sandwich in ascending order). In order to re-evaluate the chronological framework, we conducted AMS radiocarbon dating and petrographic analysis on cored peat deposits collected from near Haven Lake. Charcoal fragments collected from around Three Arm Bay, southwestern area, are also dated. This poster presents petrography and radiocarbon dates. Based on our results, eruption ages of the Main, Intermediate, Sandwich, YBO and Forty Year tephra were dated to approximately 9.5, 7.2, 4.7, 3.6 and 0.4 cal ka BP, respectively.

**Spatial and historical variation of radiocarbon marine reservoir effect around Korea****Nakanishi Toshimichi<sup>1,2</sup> Hong Wan,<sup>2,4</sup> Sung Kisuk,<sup>3</sup> Sung Kilho<sup>2,4</sup> Nakashima Rei.<sup>5</sup>**

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Since 2009, a research project to evaluate the marine reservoir effects of the coastal sites of Korea has been progressed by KIGAM AMS Lab. Estimating the reservoir effect of this area is difficult because it is hard to obtain age-known marine samples before AD 1955. In order to solve this problem, 48 sediment cores were collected with 1 m intervals by a percussion drilling tool from 61 coastal sites in the southern area of the Korean Peninsula. These drilling sites were roughly preselected by the interpretation of modern air photos of internet map services provided by the websites such as Daum(Korean Provider, [www.daum.net](http://www.daum.net)) and Google. Topographic maps in 1918-1926 with 1/50000 scale and old air photos were also used for the site selection. The length of each core was shorter than 5 m and the total drilling length was 163 m. Based on analysis of lithology and mollusk assemblages, we selected marine shell and terrestrial plant pairs from same horizontal levels. These samples were cleaned by physical and chemical pretreatments, and reduced by automatic graphitization system in KIGAM. The radiocarbon ages of the samples were measured by the 1MV AMS facility of KIGAM. These age offsets between plants and shells were correlated with the reservoir age of these sites. This presentation will also report about spatial and historical variation of radiocarbon marine reservoir effect around Korea.

**Radiocarbon ages of stalagmites from the Ryugashi Cave, Shizuoka, Japan****Tomomi Kato,<sup>1</sup> Masayo Minami,<sup>2</sup> Keiji Horikawa,<sup>3</sup> Toshio Nakamura.<sup>1</sup>**

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Stalagmite is a cave deposit precipitated from drip water. Drip water consists of carbon derived from soil CO<sub>2</sub>, which has atmospheric <sup>14</sup>C values in isotopic equilibrium with atmosphere, and carbonate-dissolved CO<sub>2</sub>, which has <sup>14</sup>C-free (dead) carbon through interaction with cave host bedrock. As a result, drip water contains some dead carbon, which will make the <sup>14</sup>C ages of the stalagmite calcite older, and so a correction of the dead carbon fraction (DCF) is needed for <sup>14</sup>C dating of stalagmites. The DCF correction is often performed on the assumption that DCF remained constant through growth time of the stalagmites. In this study, we examined the reliability of the assumption by investigating DCF in two stalagmites from the Ryugashi Cave in Hamamatsu, Shizuoka, Japan, and seasonal variation in <sup>14</sup>C concentrations of drip water in the Ryugashi Cave. The drip water samples showed an annual mean of <sup>14</sup>C of 1025±140 BP. A growing stalagmite of ~7 cm in length showed 945±30 BP at its surface and 2150±40 BP at its bottom. The calibrated age of the stalagmite was calculated by comparing the <sup>14</sup>C with the IntCal13 calibration curve, resulting that the stalagmite roughly had a constant DCF through its growth time and gives <sup>14</sup>C ages of 1050 years older than the true age. The carbon isotopic fractionation between drip water and stalagmite was negligible. The results indicate that high-resolution <sup>14</sup>C measurement can be performed on stalagmites in the Ryugashi Cave.

**AMS  $^{14}\text{C}$  dating at CIRCE. Recent cultural heritage applications****Capano Manuela,<sup>1</sup> Marzaioli Fabio,<sup>2</sup> Passariello Isabella,<sup>3</sup> D'Onofrio Antonio,<sup>4</sup> Terrasi Filippo.<sup>2</sup>**

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One of the main AMS  $^{14}\text{C}$  activities of CIRCE (Centre for Isotopic Research on Cultural and Environmental heritage) in Caserta (Italy) has always been the dating of archaeological findings and artistic monuments. This activity interested several materials (e.g. wood, bone, charcoal, seed, mortar, shell, soil, paleosoil). We present here some of the recent CIRCE analyses on cultural heritage field, performed on different kind of materials, such as mice bones from Cuma, mortars from Nola and woods from Viterbo. The bones of small mice were found in a jar in the Apollo temple of Cuma (NA - Italy). This discovery had a great impact on the archaeological community for the hypothetical link of mice with the god worship. In order to verify the antiquity of the mice, their bones were  $^{14}\text{C}$  dated. The results indicate the antiquity of mice death, occurred in the first half of IV century BC, and, at the same time, their coeval death. Mortars from the crypt of St Felix in Nola (NA - Italy) and from the basilicas of St Stephen and St Paulinus (Basilica Nova) in Cimitile (NA - Italy) were radiocarbon dated. The results confirmed the archaeological expectations, indicating the Early Christian origin of all the worship buildings, with special regard to crypt of St Felix (I-II century AD). The roof structural wooden elements of Santa Maria Nuova church in Viterbo (Italy) were dated by means of dendrochronological and radiocarbon analyses. The wiggle-matching method was applied in order to absolutely date the dendrochronological fluctuant chestnut sequences. Several building construction and renovation phases were highlighted, in agreement with the historical expectation.

## AMS-Radiocarbon dating and spectroscopic analyses of samples from Tuzapan, a Pre-Columbian archaeological site in Veracruz, Mexico

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We present the results of applying several analytical techniques, to pre-Columbian stuccos and carbon samples from Tuzapan archaeological site (Veracruz State, eastern Mexico). Carbonaceous material was found in three excavated stratigraphic levels. Radiocarbon AMS analysis and microscopic inspection identified two different types of carbonaceous materials mixed with the stucco floors : a charcoal type and a black material of granular aspect. Radiocarbon analyses of the charcoal samples gave ages of 1421 to 1640 years when all charcoal samples are considered. The granular sample gave an age older than 40,000 years. Infrared analysis of the more antique sample identified it as asphalt, confirming previous reports about the use of asphalt as a building material by antique cultures that settled in the Gulf of Mexico. The combined results from archaeomagnetic and radiocarbon dating, give a better estimation of the occupation period. Raman spectroscopy analyses determined that the stucco floor is made only by calcite and quartz, with traces of hematite and magnetite and no other extra component as pigments or minerals were detected. Information obtained by radiocarbon and archaeomagnetic analysis combined with the archaeological data contributed to complete our knowledge about the chronology of the site. Spectroscopies helped to characterize the building materials used by this culture, in order to support the archaeological studies of the region.

Topic : GAA 58

Poster Session 1

## Distribution of $^{129}\text{I}$ in terrestrial environment released from the Fukushima Daiichi Nuclear Power Plant accident

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Radioiodine is one of the most important radionuclides released from the Fukushima-Daiichi Nuclear Power Plant (FDNPP) accident.  $^{131}\text{I}$  (half-life : 8.02 days) has a short half life time. Because of the difficulty of measuring  $^{131}\text{I}$  at this time, it is expected to estimate  $^{131}\text{I}$  precipitation from  $^{129}\text{I}$  (half-life :  $1.57 \times 10^7$  years) with the long half-life in the surface soil. We have measured  $^{129}\text{I}$  concentrations in the surface soil at Fukushima.  $^{129}\text{I} / ^{127}\text{I}$  ratios were measured by accelerator mass spectrometry (AMS) at the MALT, the University of Tokyo. Stable iodine of  $^{127}\text{I}$  was determined by inductively coupled plasma mass spectrometry (ICP-MS). We already got a result that the average  $^{129}\text{I}$  concentration was  $(2.7 \pm 1.4) \times 10^8$  atoms/g prior to the FDNPP accident as  $^{129}\text{I}$  background at Fukushima. After the accident, average isotopic ratio of  $^{131}\text{I} / ^{129}\text{I}$  is estimated to  $(4.0 \pm 0.8) \times 10^{-2}$  at Fukushima as at March 11, 2011. The results of calculation about  $^{131}\text{I} / ^{129}\text{I}$  ratio made by the ORIGEN<sub>2</sub> code are  $3.2 \times 10^{-2}$  for the Unit 1 reactor,  $4.6 \times 10^{-2}$  for the Unit 2 reactor and  $4.8 \times 10^{-2}$  for the Unit 3 reactor. In this presentation, we report the distribution of  $^{129}\text{I}$  in terrestrial environment at Fukushima and  $^{131}\text{I} / ^{129}\text{I}$  ratios by region.



**Measurements of  $^{11}\text{B}/^{10}\text{B}$  isotopic ratios by AMS.**

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A campaign of characterization of nitrate pollution in rural sites in Padana valley, supported by the Italian Ministry of Agriculture, aims to establish the apportionment of various pollution sources by means of isotopic methodologies. The multi-isotope approach for studies of pollution by nitrates includes analysis of the isotopic composition of boron. Boron is a ubiquitous element, which happens to be a co-migrant of nitrate. The isotope composition of boron is not affected by nitrate conversion processes. Therefore, boron isotopes can be used to improve the identification of nitrate pollution sources in case conversion processes are involved. Boron has two stable isotopes,  $^{10}\text{B}$  and  $^{11}\text{B}$  with a natural  $^{11}\text{B}/^{10}\text{B}$  ratio of about 4 (expressed in  $\delta^{11}\text{B}$  values normalized to the standard reference material NIST 951). High precision ( $\leq 0.1\text{‰}$ ) measurements of  $^{11}\text{B}/^{10}\text{B}$  isotopic ratios are usually achieved by TIMS (Thermal Ionization Mass Spectrometry). For our purpose such a high precision is not needed because of the natural variability of the isotopic ratio in our matrices. We have then started an investigation aiming to assess if AMS (Accelerator Mass Spectrometry) allows measurements at  $\sim 2\text{‰}$  level using the CIRCE (Center for Isotopic Research on the Cultural and Environmental heritage, in Caserta) system. The purpose of this study is to integrate a new measurement method in the multi isotope approach for the determination of  $\delta^{11}\text{B}$  in water matrices. The samples will undergo a chemical purification using a selective resin, followed by measurement of the isotopic ratio  $^{11}\text{B}/^{10}\text{B}$ .

## Iodine isotopes and species in surface water transect from the North Sea to Northeastern Atlantic

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The 2010/2011 Antarctica two-ship expedition was an international scientific cruise jointly funded by the Swedish Polar Research Secretariat and the US National Science Foundation (NSF). Part of the expedition was aimed at investigating iodine isotopes ( $^{129}\text{I}$  and  $^{127}\text{I}$ ) and species of iodine ( $\text{I}^-$  and  $\text{IO}_3^-$ ) variability in surface water along transect from the North Sea through the English Channel and North Eastern Atlantic. Surface water samples were collected and analysed for total  $^{129}\text{I}$  and  $^{127}\text{I}$  isotope concentrations and their iodide and iodate species using accelerator mass spectrometry (AMS) and inductively coupled plasma mass spectrometry (ICP-MS). The results indicate a large variability in the total  $^{129}\text{I}$  and its species along the transect, whereas less change and variation is observed for the total  $^{127}\text{I}$  and its species. The highest  $^{129}\text{I}$  concentration was found in the surface water of the eastern English Channel and the lowest near to the Canary Island. Iodate seems to be the dominant iodine species for both  $^{127}\text{I}$  and  $^{129}\text{I}$  in most of the samples seawater. The results also indicate that transport of  $^{129}\text{I}$  from the western English Channel via the Biscay Bay is the main source to the observed level in the northeastern Atlantic Ocean. Another interesting feature revealed by the high  $^{129}\text{I}/^{127}\text{I}$  and distinctive  $^{129}\text{I}^-/^{129}\text{IO}_3^-$  values south of  $40^\circ\text{N}$  is the possible contribution of  $^{129}\text{I}$  through Mediterranean Outflow Water. The environmental impact of  $^{129}\text{I}$  can be a multi-axis tool, one is related to radioactivity hazards which is presently seems not harmful. The other axis is a potential tracer of water masses exchange and circulation. The third axis is a tool for ecosystem variability, where migration of fish and other marine species can be traced.

Topic : GAA 61

Poster Session 1

**Developing a Passive Zeolite Trap for Diffusive Atmospheric  $^{14}\text{CO}_2$  Sampling****Lehman Jennifer,<sup>1</sup> Xu Xiaomei,<sup>1</sup> Czimczik Claudia.<sup>1</sup>**

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Measuring  $^{14}\text{CO}_2$  of air is a powerful tool for tracing atmospheric (atm) carbon sources. Atm  $\text{CO}_2$  sampling in urban environments is challenging because of large spacial and temporal variations, and high precision  $^{14}\text{CO}_2$  monitoring is essential in remote regions for establishing a baseline for correcting community  $^{14}\text{C}$  measurements. Here, we report our progress in developing a passive zeolite trap to diffusively collect atm  $\text{CO}_2$ . The benefit of a passive diffusive atm trap is that it requires no power for sample collection. In addition,  $\text{CO}_2$  is slowly absorbed onto the sieve over a period of several days to weeks, and therefore gives a time-integrated sample of atm  $\text{CO}_2$ . Zeolite has long been used to collect environmental  $\text{CO}_2$ , by actively pumping sample gas through the sieve and more recently it has been utilized on vacuum systems to concentrate sample  $\text{CO}_2$  briefly before analysis. However, low the  $\text{CO}_2$  content of air makes sample collection using molecular sieve extremely sensitive to contamination. We tested the efficiency of cleaning and the blank of zeolite 13X. In addition we tested the memory effect by consecutively trapping and releasing modern and  $^{14}\text{C}$  deplete standard gases. The rate of  $\text{CO}_2$  absorption was monitored over several weeks and traps were tested for their maximum sampling capacity. Several different water exclusion strategies were implemented. Finally, traps were deployed to sample alongside air canisters in the field to evaluate their atm  $^{14}\text{CO}_2$  accuracy.

**Anthropogenic  $^{129}\text{I}$  in the Bering and Chukchi Seas and Arctic Ocean**

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$^{129}\text{I}$  concentrations in seawaters in the Arctic region were measured to investigate the influence of the  $^{129}\text{I}$  contaminated water discharged from nuclear fuel reprocessing facilities at Sellafield (U.K.) and La Hague (France) two decade ago [1,2]. Recently, in March 2011, a large amount of  $^{129}\text{I}$  was released to the western North Pacific by the Fukushima Daiichi Nuclear Power Plant (F1NPP) accident. To evaluate the influence of this event, we started measurements of  $^{129}\text{I}$  in seawater samples from extensive area. Seawater samples from 4 to 800 m depth at a station (74.5°N 162.0°W) in the Arctic Ocean and 9 surface water samples, from the Arctic Ocean to the western North Pacific Ocean, were collected during the R/V Mirai MR-13-06 cruise in September and October 2013, and 3 surface water samples in the Bering Sea were collected during the R/V Hakuho-Maru KH-12-4 cruise (September 2012).  $^{129}\text{I}$  in Seawater samples were purified by solvent extraction and measured by AMS at MALT, the University of Tokyo.  $^{129}\text{I}$  concentrations were as low as  $1.0\text{--}1.5 \times 10^7$  atoms  $\text{L}^{-1}$  in the western North Pacific Ocean, Bering Sea and Chukchi Sea, and increased to  $13 \times 10^7$  atoms  $\text{L}^{-1}$  in the Arctic Ocean. The distribution indicates very little influence of F1NPP accident, and also very little inflow of seawater from the Arctic Ocean to the Bering Sea. On the other hand,  $^{129}\text{I}$  concentrations for seawaters of 300-800m depth in the northernmost station were as high as  $800 \times 10^7$  atoms  $\text{L}^{-1}$ . The vertical profile indicates the presence of the Atlantic Water with high  $^{129}\text{I}$  concentration.

[1] L.W.Cooper et al. Mar. Poll. Bull. 42, 1347-1356 (2001)

[2] J.N. Smith et al. Deep Sea Res. Part I, 45, 959-984 (1998)

**AMS radiocarbon dating of early modern wooden buildings by wiggle-matching****Minoru Sakamoto,<sup>1,2</sup> Nanae Nakao,<sup>3</sup> Mineo Imamura.<sup>1,2</sup>**

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Radiocarbon dating is mainly applied for relatively ancient period and considered not to be a useful tool for recent samples. However, the accuracy of AMS radiocarbon dating has been improved, and wiggle-matching increases in the precision of the calibrated age. We carried on dating research of Japanese early modern wooden buildings in conjunction with architectural historical research. The restoration method of Japanese wood members can reconstruct the original style of the building, and wood members that were used for each time of rebuilding were selected. Tens of milligrams of tree rings were carved in 5 years each and dated by AMS. Sometimes chemicals that were used for preservation contaminate the samples, ultrasonic cleaning by solvent is good to remove them and helps to obtain the accurate radiocarbon age. It is typically the case that the total number of tree rings of wood members is less than few tens and only 3 or 4 shavings can be sampled from one member. However, their radiocarbon dates reproduce the wiggle of calibration curve well and are tolerable for discussing the detailed chronology not only the building itself but also the transition of architectural style of Japanese wooden buildings. Even if multiple candidate of calibration ages are obtained, architectural historical research can choose the proper date. Several case study of radiocarbon dating of early modern Japanese wooden buildings is shown.

**North-south transection of  $^{10}\text{Be}$  concentration in seawater in the Indian Ocean.****Yamagata Takeyasu,<sup>1</sup> Nagai Hisao,<sup>1</sup> Inoue Keisuke,<sup>2</sup> Tazoe Hirofumi,<sup>3</sup> Matsuzaki Hiroyuki.<sup>4</sup>**

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Cosmogenic nuclide  $^{10}\text{Be}$  (half-life 1.36 Ma) is produced by nuclear interaction between secondary cosmic ray and atmospheric nitrogen and oxygen. Because of  $^{10}\text{Be}$  widely spread in the ocean and the residence time is shorter than ocean-mixing time, the  $^{10}\text{Be}$  concentrations in seawater could be a useful tracer for the ocean circulation. It is necessary accumulation of oceanic  $^{10}\text{Be}$  to assess  $^{10}\text{Be}$  as a tracer for water mass. In this presentation, the north-south transection of  $^{10}\text{Be}$  concentration shows in Indian Ocean during R/V Hakuho-maru in the KH09-5 cruise, and discuss a migration of  $^{10}\text{Be}$ . The concentration of  $^{10}\text{Be}$  is high in 20-40°S and low in the south area of 40°S, which is the Antarctic Ocean, and between 20°S and 20°N in surface. According to hydrological data,  $^{10}\text{Be}$  concentration in Antarctic intermediate water (AAIW) is relatively low concentration. The AAIW come from the low concentration surface water of the Antarctic Ocean. In deep-sea, the water mass, which named Common deep water, has high concentration. The Common deep water is originated from Atlantic deep sea, but the concentration of  $^{10}\text{Be}$  in the Atlantic Ocean is lower than Common deep water. It is consider that the  $^{10}\text{Be}$  in the surface Antarctic Ocean is transported down to the Antarctic deep sea by biogenic particles in Antarctic Ocean.

**Behavior of iodine-129 and radioactive cesium in Japanese river water.**

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Radioactive iodine and cesium which emitted from the Fukushima Daiichi nuclear power plant were detected from water in environment. In the metropolitan area, higher concentration of radioactive cesium was detected from sediment of river. In this work, the long-term variations of the concentrations of the iodine-129 and radioactive cesium in the dissolved matter were observed for the Ohori River, Chiba Prefecture, and the elucidation of the actual condition was tried. We surveyed suspended soils, dissolved matter and sediment in river. The dissolved matter was obtained from the river water which was filtrated by 0.7 mm and 0.2 mm filters. The concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were obtained  $\gamma$ -ray measurement by HPGe detectors. The amounts of  $^{129}\text{I}$  were measured by AMS at MALT, the University of Tokyo. In the Showa-bashi point, the river water was sampled every two weeks at first year and monthly at second year, and the dissolved matter and suspended soils in the river water obtained the radioactive cesium and iodine-129 concentration of the moment from May, 2012. It was of observation observed that iodine-129 in dissolved matter is the concentration changes in concert with radioactive cesium, and dissolved matter (DM). Further, the river water was sampled 5-6 points in Ohori River every six months. It was showed that iodine-129 and radioactive cesium concentration change from upstream to downstream is affected by intermediate influent water.

Topic : GAA 66

Poster Session 1

 **$^{236}\text{U}$  and Pu-isotopes in two corals from French Polynesia.****Christl Marcus,<sup>1</sup> Cayeron Bruno,<sup>2</sup> Casacuberta Nuria,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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From 1966 to 1974, France conducted atmospheric nuclear bomb tests at the atolls of Mururoa and Fangataufa in French Polynesia, South Pacific Ocean. During that period, also safety experiments were performed on the coral bedrock in the North of Mururoa which generated local Pu contamination in the lagoon. To investigate the regional impact of these tests, two coral samples (*Porites A and B*) were collected on Gambier Islands about 500 km distant from the test sites for the analysis of  $^{236}\text{U}$  and Pu isotopes. This provides the first data set of  $^{236}\text{U}$  in corals from the southern hemisphere. However, due to biological activity of the so-called coral borer *lithophaga nigra* the quality of the coral samples was poor. The temporal resolution of the samples thus is only 3 to 7 years per sample with an unknown but probably large absolute age uncertainty. Our results show that the  $^{236}\text{U}$  concentrations in *Porites B* qualitatively agree with the history of atmospheric bomb testing. The  $^{236}\text{U}$  signal in *Porites A*, however, is clearly discordant in the older part of the record. Elevated  $^{236}\text{U}$  levels have been found in this part of *Porites A* (pre 1960) that could have been caused by biological induced mixing or, alternatively, that could indicate a problem with the chronology of this specimen. The results for  $^{239}\text{Pu}/^{240}\text{Pu}$  range between 0.2 and 0.05, generally agreeing with data from literature [1] but no clear trend is observed. Measured ratios indicate global fallout and local contributions of weapons grade plutonium from the safety tests, respectively.

[1] R. Chiappini et al. Sci. Total Environ. 269 (1999) 237



## Some Considerations on Radiocarbon Calibration and Confidence Intervals

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For radiocarbon dating it is necessary to calibrate the radiocarbon age obtained from the measured isotopic ratio and the radioactive decay law. The problem of radiocarbon calibration is to transform the Gaussian distribution curve of the measures radiocarbon age via a calibration curve to a probability distribution of the true calendar age of the sample. Because of the wiggly character of the calibration curve, this transformation is not a one-to-one mapping, which makes the correct determination of the true sample age and its confidence intervals a nontrivial problem that always has been a matter of discussion (see also [1]). This is not only a mathematical problem, but also a matter of the nature of the errors in radiocarbon dating. We will show some statistical paradoxes, from our point of view, of the common way to calculate the confidence intervals for the sample age and propose an alternative way and put it up for discussion.

[1] H. Dehling, J. van der Plicht, Radiocarbon 35/1 (1993) 239

## Analysis of the 1 MV CNA AMS system as a potential tool for $^{236}\text{U}$ studies in Oceanography

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The performance of 1 MV AMS system at the CNA for  $^{236}\text{U}$  measurements has been recently extensively investigated. A very promising  $^{236}\text{U}/^{238}\text{U}$  abundance sensitivity for non-processed  $^{236}\text{U}$  free samples of about  $3 \times 10^{-11}$  has been achieved. This promising result opens the door to the use of conventional low energy AMS systems to  $^{236}\text{U}$  environmental applications. The aim of this work is to determine if the sensitivity achieved for  $^{236}\text{U}/^{238}\text{U}$  is low enough to apply our system to the study of environmental processes based on  $^{236}\text{U}$  analysis. The first  $^{236}\text{U}$  results obtained on our AMS system for marine samples (sediments and water) are presented here. First, we focus on two sets of several sea-sediment aliquots from IAEA reference materials : IAEA-410, from the Bikini atoll, affected by the thermonuclear American tests; and IAEA-412, from the Pacific Ocean, presumably free from local actinides sources. Preliminary results point out to a probable  $^{236}\text{U}$  speciation between the dissolved and the particulate phases in seawater, which favours the  $^{236}\text{U}$  concentration in the sediment. We conclude that our abundance sensitivity is competitive for the study of marine sediment cores, as is further illustrated by the  $^{236}\text{U}$  results obtained on a sediment core from the Porcupine Abyssal Plain in the North Atlantic [Chamizo et al. this conference]. Second, it is discussed the  $^{236}\text{U}/^{238}\text{U}$  atomic ratio obtained on a set of 5 intercomparison sea-water samples from the North Atlantic Ocean provided by the ETH, Zürich, at the order of  $10^{-12}$ . Based on the environmental context of these samples, we discuss the versatility of the 1 MV CNA AMS and its optimization for the determination of actinides (i.e. U and Pu-isotopes). Finally, our prospects will be presented.

Topic : GAA 69

Poster Session 1

**Simulation of particle fluxes and cosmogenic nuclide production rates in meteorites****Masarik Jozef,<sup>1</sup> Beňo Juraj.<sup>1</sup>**

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The particle fluxes and production rates of cosmogenic nuclides depend on many parameters. Reliable interpretation of the measured in-situ-produced cosmogenic nuclides requires a good understanding of involved nuclear processes. We present results obtained by updated physical model for the simulation of the relevant processes. This model is enabling an investigation of nuclide production dependence on composition, shape and depth under the surface of irradiated object. LCS codes are used in our simulations for the calculation of spectra of particles inducing reactions that produce cosmogenic nuclides. Having calculated neutron fluxes with these codes, the production rates of nuclides are determined by integrating over energy the product of these fluxes with experimental and evaluated cross sections for the reaction producing particular nuclide. Dependences of production rates on chemical composition and size of irradiated object were obtained for L<sup>-</sup>, H<sup>-</sup>, CI chondrites, and iron meteorites. Both low- and high-energy products were calculated. We present also new elemental production rates calculated from our new particle fluxes and updated excitation functions. The obtained theoretical values are compared with experimental data for various meteorites. Correction factors for production rates of short-lived nuclides due to variations of galactic cosmic ray intensities were also obtained. Their importance is shown on the case of Košice meteorite.

## Estimation of the global inventory of $^{10}\text{Be}$ in soil profiles through ArcGIS

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The inventory of  $^{10}\text{Be}$ , which is deposited mainly by precipitation, in soil after its production is significant in determining the erosion rate and relative age of soils. Despite the many investigations of the relationships between  $^{10}\text{Be}$  concentrations and soil properties in a variety of places, there is a little information about estimation of the global inventory of  $^{10}\text{Be}$  in soils. We here summarize available data of  $^{10}\text{Be}$  in soils and use them for calculating the global inventory. In addition, the  $^{10}\text{Be}$  concentration is strongly influenced by grain size and a relationship between the two components can be divided into 3 categories including 12 textures on the basis of grain size according to the international soil classification system. The surface area of each of soil type can be calculated through ArcGIS. Additionally,  $^{10}\text{Be}$  in most soil profiles does not penetrate deeply, so we assume a 3m penetration depth for the calculation of inventory. Finally the difference of latitudinal averaged flux is also considered. After consideration of all these factors, we estimate the inventory of  $^{10}\text{Be}$  in global soils of different sizes. Such approach can serve as (i) reference values of the inventory of  $^{10}\text{Be}$  in global places and (ii) a start level for the development of regional models for estimation of the erosion rate and age of soils. The precision of the inventory approach can be enhanced by increasing the density of sampling sites and the exploration of soil types in future investigation.

Topic : GAA 71

Poster Session 1

**Radiocarbon measurement with 1MV AMS at charge state 1+****Sung Kilho<sup>1,2</sup> Park Gyu Jun,<sup>2</sup> Lee Jong Geol,<sup>2</sup> Hong Wan.<sup>1,2</sup>**

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A 1MV AMS was installed in KIGAM in 2007. This multi-element AMS started normal operation from January 2008 and has measured about 3000 targets annually. The AMS mainly measured  $^{14}\text{C}$  and  $^{10}\text{Be}$ , but the demand of  $^{14}\text{C}$  is higher than  $^{10}\text{Be}$ . We usually measured  $^{14}\text{C}$  at charge state 2+. At the same time, this condition always has the possibility of interference by  $\text{Li}_2$  molecular. KIGAM AMS team has treated 4130 samples including standard samples during 2013, and 258 among them were interfered by  $^7\text{Li}_2^{2+}$ . The main aim of this experiment is finding out stable condition of charge state of 1+ for age dating. At the first, beam transmission yields of ions with the charge state 1+, 2+ and 3+ were determined. The condition of stripper gas pressure was  $2.5 \cdot 10^{-2}$  mbar. For the 3+ ions, the highest yield was found to be 10.4% at 1MV terminal voltage. For the 2+ ions, beam transmission yield changed from 41.7% at 950kV to 4.7% at 200kV. For the case of 1+ ions, the highest beam transmission yield was marked 35.7% at 500kV so that terminal voltage 500kV was the finest condition for 1+ ion measurement with our machine. For the second step, stripper gas pressure was optimized for 1+ carbon ion measurement. We fixed the terminal voltage at 500kV to find out the best stripper pressure for 1+ carbon ion measurement. NIST oxalic and  $\text{CO}_2$  gas from petroleum were selected as samples.  $\text{CO}_2$  gas sample is  $^{14}\text{C}$  free material, so this sample is good to find out an ideal stripper gas pressure to break  $^{12}\text{CH}_2$ ,  $^{13}\text{CH}$  molecules effectively. The pressure of  $2.7 \cdot 10^{-2}$  mbar was most suitable to measure  $^{14}\text{C}^+$  ions at 500kV.

**$^{14}\text{C}$  dating of insects found in a glacier in Suntar-Khayata range, eastern Siberia**

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This study attempted to determine the age of organisms such as dead bees and plant fragments by radiocarbon dating from the No. 31 glacier in Suntar-Khayata range of east Siberia in order to estimate age of the glacier ice that preserved the organisms. Ice samples with organisms were collected at 0.4-1.1 m depth of five different points from the middle to the lowest part of the glacier in 2013. Radiocarbon dating of the samples was carried out using a 5 MV AMS system at the National Institute for Environmental Studies, Tandem accelerator for Environmental Research and Radiocarbon Analysis (NIES-TERRA). Each of the samples ranged from 229 to 943  $\mu\text{gC}$ . The bee found from the lowest point was estimated to be 2000 yr BP. The bees found at the higher points yielded a  $^{14}\text{C}$  age of modern age. The age for the plant fragments in the two uppermost points were 1200-1400 yr BP and in the lowest point the plant fragments were 9700 yr BP. Comparing the age of the bee with that of the plant fragments found at the same points, the plant fragments were older than the bees. Because some insects were observed in their living state on the glacier in the 2013 observation, the differences of the age indicated that the bees were alive when it arrived on the glacier. On the other hand, the plant fragments might have been already aged since detachment from the source plants. Therefore the age of the bees represented the age of the ice. This study proved a wide age gap of ice between the lowest point and the higher points. The annual mass balance observation from 2012 to 2013 showed that the melting of the ice were 2.04 m/yr at the lowest point and 1.33-1.95 m/yr at the higher points. The wide age gap of ice may be due to the difference of past melting process between the lowest and the higher points.

Topic : GAA 73

Poster Session 1

## Dating tree rings from tropical tree species by combining radiocarbon measurements and dendrochronological techniques

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In the tropics, ecosystems and populations are very vulnerable to climate change with critical implications to the food supply. Knowledge of long-term natural climatic variability is essential for understanding future changes. Dendrochronology can provide these proxies. In temperate climates, dendrochronological techniques are used to determine absolute calendar dates, but this cannot be sufficed in tropical climates. Here, we propose to use high-precision  $^{14}\text{C}$  bomb-pulse dating of selected rings to provide an independent validation of dates in tree-ring data. Two tropical species were investigated : *Prioria copaifera* (cativo) located in the Colombian Pacific region (7°15'N ; 76°58'W) and an unknown species tree from the Moraceae family collected in Bolivia (14°34'S ; 68°46'W). Approximately eight samples of each wood species were measured by  $^{14}\text{C}$ -AMS, starting before 1940 to beyond 2005 with the calendar dates provided by dendrochronological techniques. The selected rings belonged to periods before and after the  $^{14}\text{C}$  bomb spike. This allows the assessment of discrepancies between the dendrochronological dates and the  $^{14}\text{C}$  calibration curve. Our results show that growth bands of cativo were misidentified as annual rings when just a few measurements were done for the year 1995 [1]. In contrast, a remarkable agreement of the *Moracea* tree-ring/ $^{14}\text{C}$  cross-dating with the  $^{14}\text{C}$  calibration curve confirms its high potential for dendrochronological analysis. Thus, we demonstrate that few  $^{14}\text{C}$  dates after the bomb-pulse curve are not enough for dating tree rings in the tropics and alerts on the risk for misdating. [1] Jimenez & Arango 2011 Revista De Biología Tropical, 59, 1813

## The potential of iodine-129 as a hydrologic tracer

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Iodine-129 is a radioactive isotope which has long lived (half-life 15.7 Myr) and relatively mobile in the Earth's surface environment. These properties make the isotope an excellent tracer in the hydrosphere. However, use of  $^{129}\text{I}$  as a tracer in the oceans has received more attention than its use in terrestrial hydrology. We here provide a method to utilize the high sensitivity of  $^{129}\text{I}$  in order to study the potential of  $^{129}\text{I}$  as a hydrologic tracer. The aim is to expand the understanding of the complex water cycle process in a river and how can  $^{129}\text{I}$  resolve part of the problem. We collected the  $^{129}\text{I}$  concentrations of different water sources, precipitation, runoff, soil water, groundwater, and then we identified the differences of the data to find characteristics of every water source. According to these differences, we can fingerprint  $^{129}\text{I}$  as a tracer in each hydrologic system to distinguish the water sources of the river flow. We concentrate on the question of the source of  $^{129}\text{I}$ , and we built up a model with the equation of water balance and isotopic mass conservation law to distinguish the water source. Our study demonstrates that  $^{129}\text{I}$  can be a very sensitive and rather perfect hydrologic tracer. The next step is to apply this method in well specified river system in order to accurately utilize the potential of  $^{129}\text{I}$ .



**Environmental aspects of radioactive iodine in the Baltic sea region****Yi Peng<sup>1,2</sup> Ala Aldahan.<sup>3,4</sup> Göran Possnert,<sup>5</sup> Hou Xiaolin.<sup>6,7</sup>**

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The semi-enclosed Baltic Sea represents a vital economic and recreational resource for more than 90 million people inhabiting its coasts. Extensive contamination of this sea by a variety of anthropogenic pollutants has raised the concern of the people in the region. Among these pollutants is the radioactive iodine ( $^{129}\text{I}$ ), which has been emitted from a variety of sources but dominated by the marine discharges from the European nuclear reprocessing facilities. We here aim to proved environmental assessment of radioactivity hazards associated with  $^{129}\text{I}$  in the Baltic Sea. Calculations based on available concentration of the isotopes in the Baltic Sea suggest that exposure to external doses do not seem to form a serious hazard. Similarly, the situation for internal doses is apparently not hazardous where the present annual effective dose equivalent of  $^{129}\text{I}$  in human thyroid is estimated to be  $2.1 \times 10^{-8}$  Sv/y. This value is five orders of magnitude higher than pre-atmoic era level which is expected to be  $\sim 10^{-13}$  Sv/y, but three orders of magnitude less that the effective dose equivalent limit by European Nuclear society which should not exceed ( $3 \times 10^{-5}$  Sv/y). Considering the expected future increase in  $^{129}\text{I}$  accumulation in the Baltic Sea, effective dose estimation in human thyroid is also calculated on the bases of two scenarios. The first scenario considers a constant  $^{129}\text{I}$  release rate and the second is variable release rates from nuclear reprocessing facilities. In first case, the radioactivity impact is not hazardous for short time period, but the second case show that it may be a problem after 70 years. Additionally, the location of  $^{129}\text{I}$  within DNA molecules causes concern about long-term internal exposure, even at a low dose equivalent rate.

**Total inventory of  $^{129}\text{I}$  in the oceans****Chen Xuegao,<sup>1</sup> Yi Peng,<sup>1,2</sup> Ala Aldahan,<sup>3,4</sup> Yu Zhongbo.<sup>1,2</sup>**

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Applications of Iodine-129 in oceanic research have received tremendous attention during the past 20 years, and it becomes necessary to study expected hazards of  $^{129}\text{I}$  on human health and the environment. Our investigation is based on collecting data on the concentrations of  $^{129}\text{I}$  from published reports to estimate the total inventory of  $^{129}\text{I}$  in the oceans. This is performed in order to construct a comprehensive picture about the variability in the inventory of  $^{129}\text{I}$  in the oceans and analysis of regional impact on the environment and health hazards effects. We use the ArcGIS to calculate the total of  $^{129}\text{I}$ , and make two assumptions : (1) the effects of several mechanisms (such as evaporation, sedimentation, and adsorption) can be ignored and (2) the mixing of different ocean layers is negligible. Concerning the inconsistent sampling depth of  $^{129}\text{I}$ , we use imputation methods to calculate the value of every divided compartment as the input of ArcGIS. The amount of data we collected is rather uneven with respect to the hemispheres with large number in the north hemisphere and few in the southern one. This variability in data density relates to the consideration of the main emission sources of  $^{129}\text{I}$  which are concentrated in the northern hemisphere. We also discuss the reliability of results by comparing with those from pervious investigations. Although the data from recent reports are limited and inhomogeneous for accurate calculation and prediction of total  $^{129}\text{I}$  variability and environmental effects, thus we worked out a rather comprehensive data base that can be used as reference for the future research.

**Active basement uplift of the Sierra Pie de Palo case (Western Argentina)****Siame Lionel,<sup>1</sup> Michel Sébrier,<sup>2</sup> Carlos Costa,<sup>3</sup> Emilio Ahumada,<sup>3</sup> Olivier Bellier,<sup>1</sup> Bourlès Didier.<sup>1</sup>**

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The Andean back-arc of western Argentina is an obliquely converging foreland where Plio-Quaternary deformations are partitioned between strike-slip and thrust motions that are localized on the E-verging, thin-skinned Argentine Precordillera, and the W-verging thick-skinned Sierras Pampeanas, respectively. The Sierra Pie de Palo is a key structure playing a major role in the partitioning of the Plio-Quaternary deformations. Located in the westernmost Sierras Pampeanas, this mountain forms a NNE striking, 80 km-long and 35-40 km-wide, ellipsoid range that reaches elevation as high as 3162 m. It is an actively growing basement fold associated with a high level of seismic activity. To evaluate the degree of tectonic activity around the Sierra Pie de Palo, we combined a detailed morphometric analysis of the topography together with in situ-produced cosmogenic  $^{10}\text{Be}$  concentrations measured in (1) bedrock outcrops corresponding to the exhumed erosional regional surface, (2) surface boulders abandoned on alluvial fans deformed by active faults, and (3) in fluvial sediments sampled at the outlets of selected watersheds that drains out from the Sierra Pie de Palo. All together, our results allows : (1) assessing quantitative constraints on the rate of tectonic and denudation processes that are responsible for the active growth and erosion of the Sierra Pie de Palo ; (2) discussing the identification and characterization of the active faults responsible for its seismotectonic activity.

## How fast is the denudation of the Taiwan Mountains ?

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In this study, we focus on the mountains of the arc-continent collision in Taiwan, which serve as one of the best examples in the world to understand and study mountain building processes. We investigate the pattern and magnitude of denudation rates at the scale of the orogenic system, deriving denudation rates from in situ-produced cosmogenic nuclide  $^{10}\text{Be}$  concentrations measured in (1) river-borne quartz minerals sampled at major watersheds outlets, and (2) bedrock outcrops along ridge crests and at summits located along the major drainage divide of the belt. We determined a denudation pattern showing a clear discrepancy between the western ( $1.7 \pm 0.2$  mm/yr) and eastern ( $4.1 \pm 0.5$  mm/yr) sides of the range. Conversely, bedrock denudation determined along ridge crests, summits and flat surfaces preserved at high elevations are characterized by significantly lower denudation rates on the order of  $0.24 \pm 0.03$  mm/yr. Altogether, the cosmogenic-derived denudation pattern at the orogen-scale reflects fundamental mountain building processes from frontal accretion in the Western Foothills to basal accretion and fast exhumation in the Central Range. Applied to the whole orogen, such field-based approach thus provides important input data to validate and calibrate the parameters to be supplied to landscape evolution models. Moreover, the comparison between cosmogenic bedrock-derived and basin-derived denudation rates allows discussing how the topographic relief of Taiwan has evolved through the last thousands of years, and thus documenting whether or not the Taiwan Mountains are in a topographic steady state.

Topic : AHN 06

Session 7A

**Environmental Iodine-129 studies at the University of Arizona.**

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At the Arizona AMS Laboratory, we have developed a number of projects focusing on  $^{129}\text{I}$ . These studies focus on both ocean studies, particularly related to the Fukushima nuclear accident, but also to the development of  $^{129}\text{I}$  as a possible tracer of modern groundwater recharge. The Fukushima study includes monitoring of  $^{129}\text{I}$  in Pacific Ocean water samples collected on a regular basis at the Scripps Institution of Oceanography in La Jolla, CA, other coastal locations in California and also sample collection in Kaoshiung, Taiwan. This monitoring has been undertaken since shortly after the Fukushima event in 2011. Our newer study is concentrated on water in semi-arid environments. We have begun a small pilot project to demonstrate the possible usefulness of  $^{129}\text{I}$  as an age-tracer of recent recharge to shallow aquifers in the Tucson Basin. Iodine-129 concentrations have been measured in precipitation, surface water, and groundwater samples. Initial results from this study and also the on-going Pacific Ocean  $^{129}\text{I}$  study will be presented.

**Presence of  $^{236}\text{U}$  in an abyssal sediment core from the North Atlantic.**

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In this work we present the first comprehensive  $^{236}\text{U}$  data in a sediment core from the North Atlantic Ocean (4000 m depth, Porcupine Abyssal Plain (PAP) site, 49°0' N, 16°30' W). The sediment core has been further characterized through the measurement of other natural and anthropogenic radionuclides ( $^{14}\text{C}$ ,  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ). Concentration profiles show mobilization due to bioturbation, probably due to a reported increase of the seabed fauna in 1995 that followed an extra input of organic matter from the surface. Bioturbation affected differently to the different radionuclide profiles and this allowed us to go into detail on its biogeochemistry.  $^{210}\text{Pb}$  and fallout radionuclides profiles showed two secondary maxima at 2 and 6 cm depth. On the contrary  $^{236}\text{U}$  profile showed an additional pronounced maximum in most superficial layer; which may have been caused either by an additional source or by a different biogeochemical response. Similarly, a maximum was observed in the organic fraction of  $^{14}\text{C}$  which evidences the role of organic matter in the transport of  $^{236}\text{U}$ . The origin of this maxima was investigated through the analysis of the ratios of  $^{236}\text{U}$  to the other radionuclides. Finally, significantly higher  $^{236}\text{U}/^{238}\text{U}$  atomic ratios than the ones measured in the dissolved phases were obtained. This point out to a physico-chemical speciation of the antropogenic  $^{236}\text{U}$ , as previously suggested by Sakaguchi et al. [2009]. The implications of our results on the  $^{236}\text{U}$  chemistry and the potential sources of artificial radionuclides at the PAP site will be analysed. Ultimately, the potential use of  $^{236}\text{U}$  as oceanic tracer will be discussed.

**Speciation of  $^{129}\text{I}$  and  $^{127}\text{I}$  in Seawater from the Arctic.****Zhang Luyuan,<sup>1</sup> Hou Xiaolin<sup>1,2</sup> Gwynn Justin P.<sup>3</sup> Zhou Weijian.<sup>2</sup>**

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Arctic currents change and sea ice retreat have attracted much attention due to the significance on climate change, polar ecosystem and global carbon circulation. The seawater samples in upper 800 m, collected from the central Arctic Ocean during scientific expeditions of Polarstern, August-October 2011, were analyzed for chemical species of iodine isotopes. Elevated  $^{129}\text{I}$  level up to  $72.6 \times 10^8$  atoms/L was observed in the surface water in the Arctic, and a great variation from surface and deep water. Lateral and vertical exchange of water masses among Arctic components are illustrated by gradient of  $^{129}\text{I}$  concentrations in different depth, which demonstrates the Atlantic water (AW) has invaded to the Canadian Basin either across the Siberian margin seas or through the Alpha Ridge, and flows out of Arctic over the Lomonosov and Mendeleyev Ridges. Enhancement of transpolar drift (TPD) produces strong effect on the water column down to about 500 m. Iodide production is speculated to be phytoplankton dependent, and no correlation of iodine species with other water physiochemical factors (temperature, salinity, conductivity and dissolved oxygen) was observed.

**Recent evolution of  $^{129}\text{I}$  levels in the Arctic and North Atlantic Oceans.**

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$^{129}\text{I}$  is mainly released into the marine system through liquid discharge from the nuclear fuel reprocessing plants (NFRP) at Sellafield and La Hague. This radioisotope is transported around the European shelf and northwards into the Nordic Seas and Arctic Ocean. Recent studies (Casacuberta, this conference) point out to an increase of  $^{129}\text{I}$  concentrations in the Arctic during the last decade. In this work, the Irminger and Iceland Basins (IrB and IB), the transects Glasgow-Reykjavik and Sellafield-Porcupine Abyssal Plain (PAP), Norwegian and Barent Seas, Fram Strait and Greenland Sea were sampled. An outline of updated  $^{129}\text{I}$  concentrations in key locations of the North Atlantic and the Arctic Oceans is presented. An enduring increase of  $^{129}\text{I}$  concentrations throughout the North Atlantic and Arctic oceans was found, up to an order of magnitude in some areas. Our results show a weak correlation surface  $^{129}\text{I}$  - latitude, previously reported. We have evaluated if this loss might be due to the sinking of  $^{129}\text{I}$  of the Atlantic surface waters during the formation of the North Atlantic Deep Water. For that,  $^{129}\text{I}$  inventories were estimated at the Nordic Seas and IrB. They showed a strong positive correlation with latitude that might be associated to the sinking of  $^{129}\text{I}$ . Finally, high  $^{129}\text{I}$  concentrations were also detected in Southern and North-western positions (IB and PAP site); this suggests that  $^{129}\text{I}$  from Sellafield might not be exclusively transported by the North Atlantic Current and Norwegian Coastal Current into the North and Nordic Seas; the amount of  $^{129}\text{I}$  split from the main Northern branch might have implications in the evaluation of the  $^{129}\text{I}$  input function into the Arctic and its use as a water masses tracer, so further investigations would be necessary.



Topic : GAA 35

Session 7A

## Water circulation in the Norwegian Sea and the Arctic traced by AMS analysis of iodine-129.

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Two reprocessing plants at Cap La Hague (France) and Sellafield (UK) have discharged large amount of  $^{129}\text{I}$  to the sea. Based on high solubility and long residence time of iodine in seawater, reprocessing derived  $^{129}\text{I}$  was used to tracer the transport pathways and the exchange of water masses in the Norwegian Sea and the Arctic. Depth profiles of seawater were collected from 40 locations in the Norwegian Sea in 59°N -72° N in two expeditions in 2010-2011, and 60 locations in the Arctic in four Arctic expeditions during 2005-2011. 0.05-0.5 liter of seawater was taken for determination of  $^{129}\text{I}$  depending on the estimated  $^{129}\text{I}$  level in the water sample. After addition of 0.5 mg of  $^{127}\text{I}$  carrier, all iodine was first converted to iodide using  $\text{NaHSO}_3$  at pH=2 and separated by solvent extraction using  $\text{CHCl}_3$  and back extraction using  $\text{NaHSO}_3$ . The separated iodide was prepared as AgI precipitate for  $^{129}\text{I}$  measurement using accelerator mass spectrometry.  $^{127}\text{I}$  in the original seawater samples was measured using inductively coupled plasma mass spectrometry after 20 times dilution using 0.05 M ammonium. The spatial distribution of  $^{129}\text{I}$  concentrations and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in these seawater samples was drawn; and a detailed circulation pathway of water masses in the Norwegian Sea and the Arctic was derived. The whole is summarized and the major results and outcome on the circulation of the water mass in this area drawn from the  $^{129}\text{I}$  data are presented.

Topic : SP 05

Session 7A

## An approach for measurement of extremely low $^{129}\text{I}$ concentration in marine fish samples.

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Main source of  $^{129}\text{I}$  (H.L. 15.7 Myr) in the surface earth's environment today is the spent nuclear fuel reprocessing plants. In the Pacific region (far from major reprocessing plants), the anthropogenic  $^{129}\text{I}$  is transported from atmosphere into the ocean,  $^{129}\text{I}$  shows highest concentration at the surface, and then diffuses toward deeper layer (e.g. Povinec et al. 2011). This  $^{129}\text{I}$  depth profile is thus reflected by the isotopic ratio ( $^{129}\text{I}/^{127}\text{I}$ ) because  $^{127}\text{I}$  concentration is almost uniform in the seawater (ca. 60 ppb). Iodine is an essential element of marine lives. For example iodine is known to be used for metabolism action by fish. This means the  $^{129}\text{I}/^{127}\text{I}$  in marine lives should be reflected by that in seawater where the marine lives live in. However there are few reports of  $^{129}\text{I}/^{127}\text{I}$  in marine lives because  $^{129}\text{I}$  in marine lives is too rare to detect. In this study, the analytical procedure for measurement of  $^{129}\text{I}$  in fish samples using AMS (accelerator mass spectrometry) was investigated. For the extraction of iodine from fish samples, the pyrohydrolysis was applied based on the method of Schnetger and Muramatsu (1996). Samples, set in a quartz tube, were inserted into the tubular furnace gently for the fish samples not to catch fire. After the pyrohydrolysis carrier ( $^{127}\text{I}$ ) was added.  $^{129}\text{I}/^{127}\text{I}$  ratio of obtained AgI was determined by AMS.  $^{127}\text{I}$  was measured by ICP-MS. From the AMS result and the stable iodine concentration, the isotopic ratio of the fish samples themselves can be calculated. In the extraction procedure, the background (especially the cross contamination) was carefully examined by measuring blank samples repeatedly. The results of these examinations will be presented as well as the data for actual fish samples.

Topic : GAA 04

Session 7B

## Measurement of Chlorine as trace element at the ppb-level by combining neutron activation and accelerator mass spectrometry of $^{36}\text{Cl}$ .

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Neutron activation analysis using decay counting of the activated element is a well-established method in elemental analysis. However, for Chlorine there is a better alternative to measuring decay of the short-lived activation product  $^{38}\text{Cl}$  ( $T_{1/2} = 37.24$  minutes). The relatively high neutron capture cross section of  $^{35}\text{Cl}$  for thermal neutrons (43.7 barn) and the progress in the Accelerator Mass Spectrometry (AMS) technique for  $^{36}\text{Cl}$  ( $T_{1/2} = 301$  ka) allow for determination of Chlorine down to ppb-levels using practical samples sizes and standard exposure durations. For bulk solid samples a particular advantage of the method is that lab contamination can be rendered irrelevant. The  $^{35}\text{Cl}$  in the sample is activated to  $^{36}\text{Cl}$ , and surface Chlorine can be removed after the irradiation. Subsequent lab contamination, however, will not carry a prominent  $^{36}\text{Cl}$  signature. After sample dissolution and addition of sufficient amounts of Chlorine carrier the produced  $^{36}\text{Cl}$  and thus the original  $^{35}\text{Cl}$  of the sample can be determined using AMS. We have developed and applied the method for analysis of Chlorine in steel samples. The Chlorine content of steel is of interest to nuclear industry, precisely because of above mentioned high neutron capture cross section for  $^{35}\text{Cl}$ , which leads to accumulation of  $^{36}\text{Cl}$  as long-term nuclear waste. The samples were irradiated at the TRIGA Mk. II reactor of the Atominstitut in Vienna and the  $^{36}\text{Cl}$ -AMS setup of the Vienna Environmental Research Accelerator (VERA) was used for  $^{36}\text{Cl}/\text{Cl}$  Analysis.

Topic : GAA 32

Session 7B

**Measurement of  $^{36}\text{Cl}$  in surface soil around F1NPP accident site.****Miyake Yasuto,<sup>1</sup> Matsuzaki Hiroyuki,<sup>1</sup> Sasa Kimikazu,<sup>2</sup> Takahashi Tsutomu.<sup>2</sup>**

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Owing to the Fukushima Daiichi Nuclear Power Plant (F1NPP) accident, huge amount of radionuclides were released into the environment. Since the accident, very little work has been done concerning long-lived nuclides, like  $^{129}\text{I}$  ( $T_{1/2}=1.57\times 10^7\text{y}$ ) and  $^{36}\text{Cl}$  ( $T_{1/2}=3\times 10^5\text{y}$ ). These nuclides have been measured mainly by AMS and play an important role in the assessment of the effect on the environment.  $^{36}\text{Cl}$  is produced by neutron capture of  $^{35}\text{Cl}$ . In the reactors at F1NPP, there was a small amount of  $^{35}\text{Cl}$  included as impurities in the coolant.  $^{36}\text{Cl}$  have been produced during the operation until the accident and were also leaked out from the reactor like other radionuclides. In this study,  $^{36}\text{Cl}$  in surface soil collected near F1NPP were measured by AMS and results will be discussed. Two of surface soils were treated. One was collected within 10km distance from F1NPP and another was within 20km distance. Soils were homogenized and chlorine was eluted by dilute nitric acid. After removing organic matters by adsorption using activated carbon and dissolution using  $\text{H}_2\text{O}_2$ , silver chloride precipitation was obtained by adding silver nitrate solution. Finally,  $^{36}\text{Cl}$ -AMS was performed at Micro Analysis Laboratory, Tandem Accelerator (MALT), The University of Tokyo. The Chlorine isotopic atom ratios measured were  $8.7\times 10^{-11}$  and  $3\times 10^{-12}$  and these values were all above  $^{36}\text{Cl}/\text{Cl} = 4.6\times 10^{-13}$ , the atom ratio measured around F1NPP before the accident.  $^{36}\text{Cl}$  detected could be originated from the coolant in the reactor.

Topic : SP 03

Session 7B

### Optimization of the Isotope Dilution-Accelerator Mass Spectrometry (ID-AMS) technique to analyze waters with low chlorine contents.

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The cosmonuclide  $^{36}\text{Cl}$  is relevant for water researches due to its conservative chloride form.  $^{36}\text{Cl}$  measurements are commonly performed by Accelerator Mass Spectrometry (AMS) while stable chlorine concentration measurements are usually performed by ion chromatography (IC) but can also be done by Isotope Dilution-Accelerator Mass Spectrometry (ID-AMS). This last method provides several advantages: 1- the  $^{36}\text{Cl}/\text{Cl}$  ratio and the Cl concentration are measured on the same aliquot 2- the required volume of sample is reduced due to the addition of the spike 3- this method is potentially more accurate than IC.

The range of applicability of this method has been assessed on rock samples but, to our knowledge, no experimental tests were conducted on water samples characterized by low chlorine contents. This study aims at investigating the accuracy and precision of the ID-AMS method for low chlorine water samples.

The theoretical range for the measured  $^{35}\text{Cl}/^{37}\text{Cl}$  ratio yielding the lowest propagated uncertainties is between 10 and 100. All the presented results are thus within this range. Thirty-five solutions of chlorine concentrations ranging from 0.1 to 10  $\text{mg.L}^{-1}$  were prepared by weighing a diluted certified NaCl solution (Certipur Merck 99.9%) and a groundwater sample solution. Cl concentration in each solution has also been obtained by ID-AMS measurements at the ASTER 5MV-AMS facility (CEREGE, France). A linear regression between concentrations derived from both methods close to a 1:1 line ( $R^2 = 0.99$ ) is obtained. The 1-sigma uncertainties on the final chloride concentrations are lower than 5%, and follow a normal distribution. We have also theoretically estimated the optimal sample volume to reach accurate  $^{36}\text{Cl}$  and Cl measurements with ID-AMS.

**$^{236}\text{U}$ ,  $^{129}\text{I}$  and Pu-isotopes as oceanographic tracers in the Arctic and the Atlantic Ocean.**

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Recent developments in low energy AMS now allow the analysis of heavy ions including the actinides with extremely high sensitivity. As a result, the potential of using  $^{236}\text{U}$  as a new conservative and transient tracer has arisen over the last few years. In this study the first comprehensive dataset of  $^{236}\text{U}$ , spanning the Arctic and the North Atlantic Ocean is presented and the potential of combining  $^{236}\text{U}$  with  $^{129}\text{I}$  and Pu isotopes is discussed. Our results show a broad variation of  $^{236}\text{U}/^{238}\text{U}$  atom ratios, ranging from  $5\pm 5\times 10^{-12}$  in the deep Canadian Basin to  $3000\pm 90\times 10^{-12}$  in surface waters of the Eurasian Basin. These numbers correspond to the different water masses in these regions, thus proving the potential of  $^{236}\text{U}$  as an oceanographic tracer. Inventory calculations constrained the two main inputs of  $^{236}\text{U}$  to the ocean as a combination of 1000 and 1400 kg from global fallout, plus between 115 and 250 kg to the two European nuclear reprocessing plants Sellafield and La Hague. Results of  $^{129}\text{I}$  in the Arctic ranged from  $0.16\pm 0.08\times 10^7$  to  $800\pm 10\times 10^7$  at.kg<sup>-1</sup> and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios throughout the water column reflect the average global fallout ratio (i.e. 0.18). The combination of  $^{129}\text{I}/^{236}\text{U}$  ratio potentially can be used to estimate the transit time for Atlantic waters in the Arctic Ocean. Preliminary results point to transit times of from the North Sea to the Eurasian Basin, and up to 26 years to the Canadian Basin, respectively. In addition, the  $^{236}\text{U}/^{239}\text{Pu}$  ratio could be used to constrain the sources of anthropogenic radionuclides to the oceans.

Topic : FNS 4

Session 7B

**Bomb-produced  $^{236}\text{U}$  and  $^{239,240}\text{Pu}$  from a modern coral surviving the nuclear testing period at Enewetak Atoll (Marshall Islands).**Srncik Michaela,<sup>1</sup> Chan Wing,<sup>1</sup> Tims Stephen,<sup>1</sup> Fifield Keith,<sup>1</sup> Fallon Stewart.<sup>2</sup>

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At Enewetak Atoll the United States carried out an extensive series of nuclear weapon tests in the 1950's. A coral (*Porites Lutea*) growing in the western side of the atoll lagoon, between the Oak (8.9 Mt explosion yield) and Mike (10.4 Mt) test sites, was cored and processed for Accelerator Mass Spectrometry (AMS) analysis.  $^{14}\text{C}$  measurements have shown pronounced peaks in the years of testing. We have explored this further with measurements of the bomb products  $^{236}\text{U}$  and  $^{239,240}\text{Pu}$  by AMS using the 14UD pelletron accelerator at the Australian National University. The variation of both isotope concentrations as well as the  $^{236}\text{U}/^{239}\text{Pu}$  and  $^{240}\text{Pu}/^{239}\text{Pu}$  isotopic signature for the nuclear testing years is discussed. The maxima of the actinide concentrations correspond to the respective testing years. The  $^{240}\text{Pu}/^{239}\text{Pu}$  isotopic ratio shows a decreasing trend with time from 0.419 in 1952/53 to 0.086 in 1958. The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio shows a much higher variability between years and seems to be more sensitive to weapons design changes.

Topic : GAA 06

Session 7B

$^{236}\text{U}$  and  $^{239,240}\text{Pu}$  ratios from from soils around an Australian nuclear weapons test site.

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The isotopes  $^{236}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are present in surface soils as a result of global fallout from nuclear weapons tests carried out in the 1950's and 1960's. These isotopes potentially constitute artificial tracers of recent soil erosion and sediment movement. Only AMS has the requisite sensitivity to measure all three isotopes at these environmental levels. Coupled with its high throughput capabilities, this makes it feasible to conduct studies of erosion across the geographical extent of the Australian continent. In the Australian context, however, global fallout is not the only source of these isotopes. As part of its weapons development program the United Kingdom carried out a series of atmospheric and surface nuclear weapons tests at Maralinga, South Australia in 1956 and 1957. The tests have made a significant contribution to the Pu isotopic abundances present in the region around Maralinga and out to distances  $\sim 1000$  km. This would impact on the assessment techniques used in the soil and sediment tracer studies. We report recent measurements on soil samples collected from across the Maralinga Test site, and discuss the significance of these measurements at locations remote from the test area.



**17.5 Wednesday 27 August - Morning****Topic : AAT 04****Session 8****What can we learn from modeling the physics of AMS ?**

Suter Martin,<sup>1</sup> Christl Marcus,<sup>1</sup> Maxeiner Sascha,<sup>1</sup> Müller Arnold,<sup>1</sup> Seiler Martin,<sup>1</sup> Synal  
Hans-Arno,<sup>1</sup> Vockenhuber Christof.<sup>1</sup>

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The basic concepts of AMS have been known for a longtime, but are the modern instruments really optimized or could we get even better performance by using more advanced designs ? By modeling the processes which are relevant for the performance of AMS systems one gets a better understanding of the physics to answers to these questions as well as valuable tools for improving AMS setups. At the accelerator facilities at ETH many relevant processes have been studied experimentally over years and computer programs have been developed for modeling the behavior of various components. Here an overview of AMS modeling will be given and illustrated with interactive programs. The following processes will be included in the discussion : 1) The scattering processes which play a role in the stripping process and in the degrader. 2) The stopping powers which determines the peak separation in detector systems and the corresponding energy-loss straggling which affects the peak width. Together they determine the suppression power. 3) The charge state distributions and the corresponding charge changing cross sections which describe the stripping yield as function of target thickness and energy. 4) The ion optics which affects significantly the transmission and the background. Other effects such as vacuum conditions or the existence of stable molecules have to be included in optimizations procedures. Our studies show that there is a significant potential for improving AMS, but they also demonstrate the need for more reliable experimental data in order to test and refine these models.

**Isobar Separator for Anions : Current Status.**

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The Isobar Separator for Anions (ISA) for AMS is an emerging separation technique applied first to the separation of  $^{36}\text{S}$  from  $^{36}\text{Cl}$ , resulting in the relative suppression of sulphur by 6 orders of magnitude. Using a radio frequency quadrupole (RFQ) column incorporating gas cells, this innovative technique enables the use of a wide range of low energy ion-molecule reactions and collisional-induced dissociation processes for suppressing specific atomic or molecular anions with a high degree of selectivity. Beside the Cl/S pair, the other elemental pairs (analyte/isobar) successfully separated at AMS level by ISA now include Ca/K, Sr/(Y, Zr), Cs/Ba, Hf/W and Pu/U. In view of these initial successes, an effort to develop a version of the ISA that can be used as a robust technique for routine AMS analysis has been undertaken. We will present the detailed layout of a practical ISA and the functional requirements that a combined ISAAMS system should meet. These concepts are currently being integrated into a pre-commercial ISA system that will be installed soon at the newly established A. E. Lalonde AMS Laboratory in Ottawa, Canada

**Isobar-separation techniques for 6 MV Tandem accelerators.****Vockenhuber Christof,<sup>1</sup> Miltenberger Klaus-Ulrich,<sup>1</sup> Suter Martin,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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Middle-sized tandem accelerators with terminal voltages around 6 MV are wide-spread among the AMS facilities. The ETH Zurich 6 MV EN tandem, in operation since 1964, was one of the first accelerators that was converted to an AMS system and subsequently measurement methods have been developed for all classical AMS nuclides ( $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{129}\text{I}$ ). Today the role of these tandems in AMS is changing because most of these nuclides can be measured with much less effort at low-energy AMS facilities. Only nuclides that require suppression or identification of a stable isobar (e.g.  $^{36}\text{Cl}$ ) require higher energies available at larger facilities. There are even more nuclides in the medium mass regime (e.g.  $^{32}\text{Si}$ ,  $^{53}\text{Mn}$ ,  $^{59}\text{Ni}$ ,  $^{60}\text{Fe}$ ) that are currently the domain of the larger tandems ( $>10$  MeV). It is not clear yet if those are measurable at energies available at 6 MV tandem accelerators. With the advances of detector systems like ionization chambers at low energies or the use of highly homogeneous and robust silicon nitride foils as degrader foils or entrance windows one can expect some progress in the isobar separation at least for some of these nuclides. We will review the isobar-separation techniques that are available at 6 MV tandem accelerators and show with some examples what are the physical limits of these techniques.

Topic : AAT 07

Session 8

**The ILIAS project for selective isobar suppression by Laser photodetachment.**

**Forstner Oliver,<sup>1,2</sup> Andersson Pontus,<sup>3</sup> Hanstorp Dag,<sup>4</sup> Lahner Johannes,<sup>1</sup> Martschini Martin,<sup>1</sup> Pitters Johanna,<sup>1</sup> Priller Alfred,<sup>1</sup> Steier Peter,<sup>1</sup> Golser Robin.<sup>1</sup>**

[1]VERA Laboratory, Faculty of Physics, University of Vienna (Austria)

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[3]Department of Earth and Space Sciences, Chalmers University of Technology (Sweden)

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Laser photodetachment is the process of removing the extra electron of a negative ion by means of laser radiation. This can happen only if the photon energy is higher than the electron affinity of the investigated ion. This process can be used in mass spectrometry to selectively suppress unwanted isobars provided that the electron affinity of the unwanted isobar is lower than the isobar under investigation. The ILIAS setup (Ion Laser InterAction Setup) at the University of Vienna has been constructed to study laser photodetachment of negative atomic and molecular ions and evaluate its applicability for selective isobar suppression in accelerator mass spectrometry. It provides mass separated beams of negative ions with energies up to 30 keV. The negative ions are produced in a Middleton type cesium sputter ion source, mass selected and slowed in a gas-filled radio frequency quadrupole cooler where they are overlapped with a strong continuous wave laser beam. By careful selection of the photon energy only unwanted isobars are neutralized while the isobar of interest remain negatively charged. A detailed description of the setup will be presented. Thereafter, the first photodetachment experiments of atomic and molecular ions with the RFQ cooler will be presented. Special focus will be given on mass systems relevant for AMS. Furthermore, a possible scheme for application of this new method to a 3 MV AMS facility will be described.

**Studies of the intrinsic ion transmission of RF ion guides for AMS : I.****Zhao Xiaolei,<sup>1</sup> Litherland Albert.<sup>2</sup>**

[1]Dept. of Physics and A. E. Lalonde Lab, Univ. of Ottawa, (Canada)

[2]Department of Physics, University of Toronto, (Canada)

The use of a radio frequency quadrupole (RFQ) controlled gas cell to facilitate on-line isobar separations for accelerator mass spectrometry (AMS) has been a subject of on-going exploration recently. There are many new technical challenges to adapt an RF gas cell device for routine AMS and still maintain high and stable ion transmission efficiency. In this simulation study using mainly SIMION 8.1, but also with some theoretical computations involving Mathieu functions to check the simulation accuracy for the hyperbolic electrode RFQ, we present a systematic assessment of the intrinsic abilities of linear RFQ ion guides to transmit ions in vacuum. This can be done with high accuracy by considering first an axially symmetric ion beam that begins its motion inside an ideal (infinitely long) RFQ ion guide consisting of hyperbolic electrodes. The practical relevance of the results obtained will be discussed. The basic finding is that anions in the phase space typically used in AMS can readily be accommodated in a practical RF ion guide in vacuum. However, this statement lead to several directions for the continuing studies that include practical concerns such as the optimum way to get the ions in and out of the gas-filled RF ion guide, and how to assess and control the dynamic range and distribution of the ions' instantaneous kinetic energies throughout the region of gas interactions. These will be briefly discussed also, but the main conclusion of the present work supports the development of highly efficient RF ion guide gas cell devices using standard ion sources for routine AMS.

Topic : AAT 08

Session 9A

**Isobar separation of  $^{93}\text{Zr}$  and  $^{93}\text{Nb}$  at 24MeV with a new multi-anode ionization chamber.**

Martschini Martin,<sup>1</sup> Buchriegler Josef,<sup>1,2</sup> Collon Philippe,<sup>3</sup> Kutschera Walter,<sup>1</sup> Lachner Johannes,<sup>1</sup> Lu Wenting,<sup>3</sup> Priller Alfred,<sup>1</sup> Steier Peter,<sup>1</sup> Golser Robin.<sup>1</sup>

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$^{93}\text{Zr}$  with a half-life of 1.6 Ma is produced with high yield in nuclear fission, and thus should be present as a natural or anthropogenic trace isotope in all compartments of the general environment. This isotope would immediately find numerous applications, however, its detection at sufficiently low levels has not yet been achieved. AMS measurements of  $^{93}\text{Zr}$  suffer from the interference of the stable isobar  $^{93}\text{Nb}$ . At the Vienna Environmental Research Accelerator VERA a new multi-anode ionization chamber was built. It is optimized for isobar separation in the medium mass range and is based on the experience from AMS experiments of  $^{36}\text{Cl}$  at our 3MV-facility. The design provides high flexibility in anode configuration and detector geometry. After validating the excellent energy resolution of the detector with  $^{36}\text{Cl}$ , it was recently used to study Iron-Nickel and Zirconium-Niobium-Molybdenum isobar separation. To our surprise, the separation of  $^{94}\text{Zr}$  ( $Z=40$ ) from  $^{94}\text{Mo}$  ( $Z=42$ ) was found to be much better than that of  $^{58}\text{Fe}$  ( $Z=26$ ) from  $^{58}\text{Ni}$  ( $Z=28$ ), despite the significantly larger  $\Delta Z/Z$  of the latter pair. This clearly contradicts results from SRIM-simulations and suggests that differences in the stopping behavior may unexpectedly favor identification of  $^{93}\text{Zr}$ . At 24 MeV particle energy, a  $^{93}\text{Nb}$  ( $Z=41$ ) suppression factor of 1000 was achieved based on a  $^{93}\text{Zr}$  spectrum obtained by interpolation between experimental spectra from the two neighboring stable isotopes  $^{92}\text{Zr}$  and  $^{94}\text{Zr}$ . Assuming realistic numbers for chemical Niobium reduction, a detection level of  $^{93}\text{Zr}/\text{Zr}$  below  $10^{-9}$  seems feasible.

Topic : FNS 1

Session 9A

**Development of  $^{93}\text{Zr}$  - Nb separation for future AMS measurement.**

**Lu Wenting,<sup>1</sup> Collon Philippe,<sup>1</sup> Steier Peter,<sup>2</sup> Kashiv Yoav,<sup>1</sup> Ostdiek Karen,<sup>1</sup> Bauder William,<sup>1</sup>  
Skulski Michael,<sup>1</sup> Anderson Tyler,<sup>1</sup> Lachner Johannes,<sup>2</sup> Martschini Martin,<sup>2</sup> Bowers Matt.<sup>1</sup>**

[1]University of Notre Dame (United States)

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Knowledge of  $^{93}\text{Zr}$  and other stable Zr isotopes neutron-capture cross-sections has relevance in both nuclear astrophysics related questions and nuclear waste management.  $^{93}\text{Zr}$  is mostly produced in the s-process in AGB stars, at the meeting point of the main and weak s-process. Large uncertainty exists in current  $^{93}\text{Zr}$  Maxwellian Average cross section while the model study requires more accurate values. Large amounts of  $^{93}\text{Zr}$  are produced in the nuclear reactor, either as a fission product or as the result of neutron capture on stable Zr isotopes. Considering its long term radiological effect, great efforts have been done on its transmutation study. Again, accurate Zr isotopes neutron capture cross section is required for this purpose. Accelerator Mass Spectrometry (AMS) is ideally suited to study this question. Activation+AMS can be used to measure  $^{92}\text{Zr}(n,\gamma)^{93}\text{Zr}$  value. Also,  $^{93}\text{Zr}$  measurement by the AMS method can be utilized to determine  $^{93}\text{Zr}$  level present in nuclear waste for nuclear waste management purpose. At the Nuclear Science Laboratory at University of Notre Dame, we are developing an AMS technique to measure  $^{93}\text{Zr}$ . The major difficulty is the separation of  $^{93}\text{Zr}$  from its stable isobar  $^{93}\text{Nb}$ . Currently we are working with the gas-filled magnet technique in combination with gas ionization chamber. A procedure to chemically reduced Nb content has been developed and tested. The reduction factor has been measured to be a factor of 1000 through AMS method done at VERA, University of Vienna.

Topic : AHN 09

Session 9A

**Determining  $^{210}\text{Pb}$  by accelerator mass spectrometry****Sookdeo Adam,<sup>1</sup> Cornett Jack,<sup>1</sup> Zhao Xiaolei,<sup>2</sup> Charles Christopher,<sup>2</sup> Kieser William.<sup>2</sup>**

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Beams of  $\text{PbF}^{3-}$  ions were produced initially using the 834 ion source at IsoTrace with targets of  $\text{PbF}_2$ . Since a large count rate of common Pb could interfere with measurement of the rare  $^{210}\text{Pb}$  isotope we examined alternative target preparation chemistry to produce  $^{210}\text{PbF}^{3-}$  beams. Beams of 150 to 175 nA of  $^{208}\text{PbF}^{3-}$  were measured in targets prepared by adding the Pb to  $\text{HF}(\text{aq})$  and equal parts of  $\text{CsF}$  and  $\text{AgF}_2$  and dried in a clean Savillex container. Although, this ion beam produces the highest counts of  $^{210}\text{Pb}$ , the factors controlling the beam stability require further study.  $^{210}\text{Pb}$  was detected in the +3 charge state using a conventional gas ionization detector at the IsoTrace Facility. Interference from the sum peak of  $^{70}\text{Zn}$  and  $^{140}\text{Ce}$  was measured in some targets. An anion exchange column separation was developed to separate  $^{210}\text{Pb}$  from  $^{70}\text{Zn}$  and  $^{140}\text{Ce}$ . Using this technique the sum peak of  $^{70}\text{Zn}$  and  $^{140}\text{Ce}$  was almost completely eliminated. We tested two different approaches to quantify the  $^{210}\text{Pb}$  concentration : (1) Measuring the  $^{210}\text{Pb} : ^{205}\text{Pb}$  ratio after adding 7.2pg of  $^{205}\text{Pb}$ , and (2) Measuring the  $^{210}\text{Pb} : ^{208}\text{Pb}$  ratio after adding 100 $\mu\text{g}$  of  $^{208}\text{Pb}$  and using a calibration curve coupled with the measurement of the concentration of  $^{208}\text{Pb}$  by ICPMS. The measurement of  $^{205}\text{Pb}$  was difficult because of  $^{205}\text{Tl}$  interference and molecular interferences at mass 205. Using the second technique, initial measurements of  $^{210}\text{Pb}$  in the CLV1 standard reference material agreed with the certified value of 660mBq  $\text{g}^{-1}$ . Further work, using the high resolution injection magnet at the A. E. Lalonde AMS Laboratory at the University of Ottawa will improve the precision of these measurements.



Topic : GAA 20

Session 9A

**The distribution of  $^{236}\text{U}$  and  $^{129}\text{I}$  in the North Sea in 2009.**

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Goroncy Ingo,<sup>2</sup> Herrmann Jürgen.<sup>2</sup>

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The North Sea is of particular interest for tracer studies because it represents the source region for many anthropogenic nuclides released by the two European nuclear reprocessing facilities located in Sellafield (GB) and La Hague (F). Tracers released from these facilities are mixed in the North Sea before they spread into the Arctic Ocean together with the northeastward flowing Atlantic water masses. Previous studies have shown that radioactive tracers released into the North Sea region can be used to determine tracer transit time distributions in the Arctic Ocean [1]. Our recent studies suggest that the ratio of  $^{129}\text{I}/^{236}\text{U}$  could be well suited as a new tool to determine transit times of Atlantic water masses in the Arctic Ocean. To use this ratio as a proxy for the transit time of water masses the input function has to be well known. In this study the  $^{129}\text{I}/^{236}\text{U}$  ratio was measured in 40 samples collected in the North Sea in 2009. All analyses were performed on the Compact AMS system Tandy at ETH Zürich. The measured concentrations and the isotopic ratios agree well with the documented releases of  $^{129}\text{I}$  and  $^{236}\text{U}$  from Sellafield and La Hague into the North Sea region. Further, a simple conceptual model is used to calculate the tracer input function for the Arctic Ocean. Our results show that both, the measured tracer concentrations and the  $^{129}\text{I}/^{236}\text{U}$  ratios agree well with the modeled values, thus providing an important tool for the determination of transit times of Atlantic waters in the Arctic Ocean.

[1] J. N. Smith, F. A. McLaughlin, W. M. Smethie, S. B. Moran, K. Lepore, Iodine-129,  $^{137}\text{Cs}$ , and CFC-11 tracer transit time distributions in the Arctic Ocean. J.G.R. : Oceans 116, C04024 (2011).

Topic : PRE 09

Session 9A

**Measurement  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$  by accelerator mass spectrometry at CIAE.**

He Ming,<sup>1</sup> Xu Yongning,<sup>1</sup> Du Liang,<sup>1</sup> Yang Xuran,<sup>1</sup> Wang Xiaoming,<sup>1</sup> Zhao Qingzhang,<sup>1</sup> Jiang Shan,<sup>1</sup>  
Cai Li,<sup>1</sup> Lan Xiaoxi,<sup>1</sup> Pang Fangfang,<sup>1</sup> Wu Shaoyong,<sup>1</sup>

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The long lived isotopes of  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$  can be used in many applications including radioactive wastes magnet, neutron dosimetry, cosmic radiation and so on. Based on the large accelerator (terminal voltage 12MV) and a big Q3D magnet spectrometry the measure method for determination  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$  are being developed at the AMS facility at China Institute of Atomic Energy (CIAE). In order to achieve the sensitivity required for such applications, the following techniques for removing  $^{59}\text{Co}$  and  $^{63}\text{Cu}$  interference in the case of  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$  measurement have been developed. (1) Chemical procedures were developed for removing Co and Cu in the sample, the content of Co and Cu in the sample can be decrease to less than 50ppm after the chemical procedures. (2) high purity Al target holders were used instead of NEC target holder to decrease the background level of Co and Cu. This is also important factor for decreasing the background. (3)an  $\Delta E$ -Q3D system which consist of a Q3D magnetic spectrometer with absorber at its entrance was used, Due to the different energy losses of isobars in the absorber, the  $^{59}\text{Ni}$  and  $^{59}\text{Co}$ ,  $^{63}\text{Ni}$  and  $^{63}\text{Cu}$  were separated at the focal plane of the Q3D magnet spectrometer. Suppression factor of about 103 were achieved for isobar ions.(4) A four anode gas ionization chamber was set at suitable position of the Q3D focal plane to further identify  $^{59}\text{Ni}$  and  $^{59}\text{Co}$  or  $^{63}\text{Ni}$  and  $^{63}\text{Cu}$ . Based on these works, the detect sensitivity of  $^{59}\text{Ni}$  and  $^{63}\text{Ni}$  are  $3 \times 10^{-13} (^{59}\text{Ni}/\text{Ni})$  and  $6 \times 10^{-13} (^{63}\text{Ni}/\text{Ni})$  respectively.

Work supported by the National Science Foundation of China, under Grant No. 11175266.

Topic : AAT 06

Session 9B

**Radiocarbon positive-ion accelerator mass spectrometry.****Freeman Stewart,<sup>1</sup> Shanks Richard,<sup>1</sup> Donzel Xavier,<sup>2</sup> Gaubert Gabriel,<sup>2</sup>**

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Positive-ion accelerator mass spectrometry of natural-abundance  $^{14}\text{C}$  is demonstrated. Positive carbon ions are extracted from a Pantechnik Nanogan electron cyclotron resonance ion source and injected onto the deck of the SUERC bipolar single-stage accelerator mass spectrometer for ion identification. This is proof-of-principle of an alternative to the conventional negative-ion radiocarbon measurement scheme, with different limitations.

**Status on mass spectrometric radiocarbon detection at ETHZ.****Seiler Martin,<sup>1</sup> Maxeiner Sascha,<sup>1</sup> Wacker Lukas,<sup>1</sup> Synal Hans-Arno,<sup>1</sup>**

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Initial experiments have demonstrated the feasibility of radiocarbon detection with a tandem mass spectrometer (MS) utilizing negative ions extracted at 45 keV from a sputter ion source, destroying any molecular interferences in a He gas stripper, and analyzing atomic ions in a second mass filtering stage built from magnetic and electrostatic filters [Synal et al. 2013]. This system did not include a second acceleration step and thus, can be regarded as the first true MS system for radiocarbon detection.

At ETHZ we have finalized an experimental platform to optimize the performance of such a system. A key point is the phase space measurement of the ETH ion source. Because of the low energy of the ions injected into the stripper canal, a much lower phase space compression is achieved, requiring a larger acceptance of the stripper tube to avoid significant ion beam losses. The gas flow leaking out of the stripper canal into the second spectrometer part is a source of background events. The dependency of the background on the pressure in the electrostatic analyzer was measured showing limits for the stripper size through its gas conductivity. Another important point is the detection efficiency for  $^{14}\text{C}$  ions. For counting the single ions an electron multiplier tube was used. Efficiencies in the given energy range and lifetime in the instrument will be discussed. The performance of the latest setup will be shown, discussing sample throughput, background level and measurement efficiencies as well as possible applications.

Topic : AAT 16

Session 9B

**Quantification of  $^{14}\text{C}$  with Cavity Ring-Down Spectroscopy.****McCartt Alan,<sup>1</sup> Ognibene Ted,<sup>1</sup> Bench Graham,<sup>1</sup> Turteltaub Kenneth.<sup>2</sup>**

[1]Center for Accelerator Mass Spectrometry (CAMS), Lawrence Livermore National Laboratory, (United States)

[2]Biology and Biotechnology Division, Lawrence Livermore National Lab (United States)

Accelerator Mass Spectrometry (AMS) is currently the most sensitive method for quantitation of  $^{14}\text{C}$  in biological samples. This technology has been used in a variety of low dose, human health related studies over the last 20 years where very high sensitivity is needed. AMS pioneered these scientific methods, but its expensive facilities and requirements for highly trained technical staff have limited their proliferation. Quantification of  $^{14}\text{C}$  by cavity ring-down spectroscopy (CRDS) offers an approach that eliminates many of the shortcomings of an accelerator-based system and would supplement the use of AMS in biomedical research. Our initial prototype, using a non-ideal wavelength laser and under suboptimal experimental conditions, has a 20-modern limit of detection in milligram-sized samples. These results demonstrated proof of principle and provided the specifications needed for a system that would have  $^{14}\text{C}/\text{C}$  sensitivity in the  $1 \times 10^{-13}$  range. Presented here are the results of our new system, which consists of a completely rebuilt cavity, ideal wavelength laser, and ultra-cold test gas temperatures.

Work performed at the Research Resource for Biomedical AMS, which is operated at LLNL under the auspices of the U.S. Department of Energy under contract DE-AC52-07NA27344, and is supported by the National Institutes of Health (NIH), National Institute of General Medical Sciences (NIGMS), Biomedical Technology Research Resources (BTRR) under grant number 8P41GM103483.

Topic : GAA 02

Session 9B

**Developments in ramped-combustion radiocarbon analysis of natural sediment :  
towards correcting organic carbon composition ( $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$ ) for carbonate  
contribution.**

**Soulet Guillaume,<sup>1</sup> Jordon D. Hemingway,<sup>1,2</sup> Ann P. McNichol,<sup>1</sup> Valier Galy.<sup>1</sup>**

[1] Woods Hole Oceanographic Institution (United States)

[2] Massachusetts Institute of Technology (United States)

Ramped-combustion and ramped-pyrolysis radiocarbon analysis have been recently used to separate sedimentary organic matter based upon relative reactivity. This approach improves our understanding of the age and reactivity structure of sedimentary organic carbon with applications in carbon cycle science and geochronology [1, 2]. Published analyses were performed on carbonate-free sediment after acid treatment. However, acidification was shown to reorganize the organic matter reactivity structure [3] potentially complicating interpretation of the reactivity and isotope distribution. Here, we investigate whether the age and reactivity spectra of organic matter can be inferred through ramped-combustion of raw (i.e. non acidified) sediment samples from various environments (ocean, lake, river, soil). Qualitative observations of the analyzed samples suggest that maxima in eluted  $\text{CO}_2$  peak at predictable temperatures. For samples characterized by a large proportion of carbonates,  $\delta^{13}\text{C}$ - $\Delta^{14}\text{C}$  data from the collected  $\text{CO}_2$  fractions display a mixing line between the "cold-organic" and the "hot-carbonate" end-members. Thus, we developed a two step methodology to back calculate the "actual" isotopic values of organic and carbonate components : 1) the sample thermogram is deconvolved through Gaussian - skew Gaussian decomposition and, 2) a Monte Carlo approach is applied to the isotopic mass balance of each collected  $\text{CO}_2$  fraction. Results were compared to artificial control samples to investigate the applicability of the approach to raw (i.e. non acidified) natural samples.

[1] Rosenheim and Galy (2012), Geophys. Res. Lett. 37, L19703

[2] Rosenheim et al. (2013), Radiocarbon 55(1), 115-126

[3] Plante et al. (2013), Radiocarbon 55(2), 1077-1083

Topic : SP 10

Session 9B

**Second generation laser-heated microfurnace for graphitisation of microgram samples.****Yang Bin,<sup>1</sup> Smith Andrew,<sup>1</sup> Long Shane.<sup>1</sup>**

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Based on our systematic studies of the prototype microfurnace (MF-I), the second generation laser-heated microfurnace (MF-II) has been made with the following features : 1) it has a small reactor volume of 0.25 cc allowing us to completely graphitise carbon dioxide samples containing as little as 2  $\mu\text{g}$  of carbon, 2) it can operate over a large pressure range (0 to 3 bar) and so has the capacity to graphitise  $\text{CO}_2$  samples containing up to 100  $\mu\text{g}$  of C ; 3) it is compact, with three valves integrated into the microfurnace body, 4) It permits future automation of the process, 5) it is compatible with our new version of small-sample conventional graphitisation furnace. We use a "budget" fibre packaged array for the diode laser with custom built focusing optics. The use of a new infrared (IR) thermometer with a short focal length has allowed us to decrease the height of the unit. These innovations have produced a cheaper and more compact device and two identical units have been constructed and tested. Feedback control of the catalyst temperature and logging of the reaction parameters is managed by a LabVIEW interface. To quantify the extraneous carbon added to the sample during graphitisation, a series of  $\text{CO}_2$  samples ranging in size from 2 to 40  $\mu\text{g}$  of carbon were prepared  $^{14}\text{C}$ -depleted  $\text{CO}_2$  were measured by ANTARES. Both versions of MF-II behaved similarly, with 0.050 $\mu\text{g}$  and 0.045 $\mu\text{g}$  extraneous carbon added, assuming an activity of 100 pMC.

**17.6 Wednesday 27 August - Afternoon****Topic : PRE 04****Session 10A****Upgraded isotope-cycling system for the 14UD Pelletron accelerator at the Australian National University.**

**Fifield L. Keith,<sup>1</sup> De Cesare Mario,<sup>1</sup> Weisser David,<sup>1</sup> Cooper Alan,<sup>1</sup> Tsifakis Dimitrios,<sup>1</sup> Tims Stephen,<sup>1</sup> Wallner Anton,<sup>1</sup> Lobanov Nikolai,<sup>1</sup> Tunningley Thomas,<sup>1</sup> Heighway Justin,<sup>1</sup> Bockwinkel John.<sup>1</sup>**

[1]Australian National University (Australia)

AMS takes ~30% of the beam time on the 14UD Pelletron accelerator at the Australian National University. The remainder is divided between fundamental nuclear physics (50%) and materials (20%) research. Due to the imperatives of operating in this shared environment, cycling between isotopes requires changing the field in the injection magnet, and hence is "slow". Switching times are ~10s, and cycle times typically 300s. This has limited the precision that can be obtained for AMS measurements to ~3%. In order to improve precision and automation, a fast-cycling system is, however, under development. This system has some novel features at the low-energy end. Because there is a fast beam-chopper immediately after the magnet, and the magnet must rotate between ion sources, the conventional option of "bouncing" the insulated vacuum box of the magnet was not feasible. Instead, a  $\pm 10$  kV TREK fast high-voltage amplifier in series with our -200kV Glassman pre-acceleration power supply is used to change the energies of the beams after the ion source. This implies, however, that the different beam energies are maintained all the way to injection into the accelerator. Hence, the electrostatic quadrupole triplet lens that focuses and steers the beams into the accelerator must also be changed from beam to beam. The focusing and x-y steering capability of this lens therefore requires a further six TREK 0-15 kV fast high-voltage amplifiers. At the high energy end, a new vacuum box has been installed in the analysing magnet, which widens substantially along its length in order to accommodate the off-axis beams. It is followed by a new box that houses the off-axis cups. This new fast-cycling system will be tested over the coming months, and we will report on progress.



Topic : PRE 08

Session 10A

**AMS with heavy nuclides at the Munich Tandem accelerator.**

Korschinek Gunther,<sup>1</sup> Faestermann Thomas,<sup>1</sup> Fimiani Leticia,<sup>1</sup> Gómez Guzmán José Manuel,<sup>1</sup> Hain Karin,<sup>1</sup> Ludwig Peter.<sup>1</sup>

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We have extended and optimized our AMS set-up at the Munich MP Tandem ( $TV \approx 13$  MV). Radionuclides, mainly of masses heavier than  $\approx 36$  are within the scope of our interests because of their high scientific potentials which can be studied by the unique sensitivity of a large facility. As a large isobaric background is in many cases a strong limitation we are using since many years a gas-filled magnet and a multi dE ionization chamber for its suppression. Two preceding Wien filters and a time-of-flight path allow a further suppression of non-isobaric background in this beam line. The optimization of the setup as a whole yields quite low detection limits, for example of  $^{53}\text{Mn}/\text{Mn} \approx 2 \times 10^{-15}$ , and  $^{60}\text{Fe}/\text{Fe} \leq 1 \times 10^{-16}$ , while the overall efficiency (including ion source) is typically  $10^{-5}$ - $10^{-4}$ . A second beam line serves for very heavy nuclides like the actinides. Here, after the two Wien-filters, a third Wien-filter with a high mass resolution of around 1/120 and a succeeding time-of-flight path help for a drastic reduction of any background. Depending on the isotope different detectors may terminate this beam line. Common for both lines is a negative ion injector with a mass resolution of around 1/400 and the use of different dedicated ion sources depending on the isotopes of interest. Besides the discussion of the set-up also ongoing measurements will briefly be addressed.

Topic : SP 09

Session 10A

## Electrodeposition as an alternate method for preparation of environmental samples for iodide for analysis by AMS.

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We present an evaluation of an alternate method for preparing environmental samples for  $^{129}\text{I}$  analysis by Accelerator Mass Spectrometry (AMS) at the Idaho National Laboratory. The optimal sample preparation method is characterized by ease of preparation, capable of processing very small quantities of iodide, and ease of loading into a cathode. Electrodeposition of iodide on a silver wire was evaluated using these criteria. The results of this study indicate that the electrochemically formed silver iodide deposits produce ion currents similar to those from precipitated silver iodide. Electrodeposition allows the processing of sub-microgram quantities of iodide. The major advantage of this method is that the silver wire/electrodeposited silver iodide is much easier to load into a cathode. Precipitated silver iodide samples are usually mixed with niobium or silver powder prior to loading in a cathode. Using electrodeposition, the silver is already mixed with the sample and can simply be picked up with tweezers, placed in the sample die and pressed into a cathode.

Topic : AAT 09

Session 10A

## Preliminary measurements on the new TOF system installed at the AMS beam line of INFN-LABEC.

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A high resolution Time of Flight (TOF) system has been developed at LABEC, the 3 MV Tandem accelerator laboratory in Florence, in order to improve the sensitivity of AMS measurements on carbon samples with ultra-low concentration and to also measure other isotopes, such as  $^{129}\text{I}$ . The system can be employed to detect and identify residual interfering particles originated from the breakup of molecular isobars. The set-up has been specifically designed for low energy heavy ions : it consists of two identical time pick-off stations, each constituted of a thin conductive foil and a Micro-Channel Plate (MCP) multiplier. The beam line is also equipped with a silicon detector, installed downstream the stop TOF station. Here, we report on the design, the readout electronics and the preliminary measurements of the new system. Tests on the single timing station allowed us to evaluate the maximum contributions of the electronics and the intrinsic MCP resolution to the uncertainty of the timing measurement (FWHM 00 ps). To this purpose, single particle pulsed beams of 2-5 MeV protons and 10 MeV  $^{12}\text{C}^{3+}$  ions, to simulate typical AMS conditions, were used. Preliminary TOF and TOF-E measurements were also performed after the installation of the system on the AMS beam line with carbon beams, using the foil-MCP and the silicon detector as the start and the stop signals, respectively. The acquired spectra suggest a small residual background from neighbouring masses reaching the end of the beam line with the same energy as the rare isotope.

## AMS method for depth profiling of trace elements concentration in materials - construction and applications.

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From its advent more than 30 years ago accelerator mass spectrometry was involved in a tremendous effort for covering the social demands of such sensitive applications. The need for investigation of material behavior to the impact/retention/repulsion/ contamination with dangerous or radioactive elements has driven the AMS to the development of a modified analyzing method : the AMS depth profile method (AMS-DPM) [1-4]. It measures continuously the concentration of a trace element in a given sample material as a function of depth from the surface (eg. T in C, D in W etc). To perform DP a common AMS facilities has to undergo several changes : a new replaceable sample target has to be constructed that accepts large, square size samples. Its position has to be adjusted in focus point of the sputter beam. Crater rim effects of the produced hole in the sample have to be avoided or removed from the registered events in the detector. Suitable reference samples have to be prepared and used for calibration. All procedures will be presented in the paper. Also, applications will be presented concerning actual problems of construction of new types of protection tiles for the reaction vessels in fusion reactors.

[1] C. Stan Sion, R. Behrisch, J. P. Coad, U. Kreissig, F. Kubo, V. Lazarev, S. Lindig, M. Mayer, E. Nolte, A. Peacock, R. Rohrer, J. Roth , J.Nucl. Mat.290-293 (2001)491-495

[2] Stan-Sion, L. Rohrer, P. Hartung , V. Lazarev, R.Luther, E. Nolte, R. Behrisch, J. Roth, NIM B192,3 (2002), 331-338

[3] M. Enachescu, V. Lazarev and C. Stan-Sion, J. Phys. D : Appl.Phys, Volume 39, (2006) 2876-2880

[4] C. Stan-Sion, M. Enachescu, O. Constantinescu, M. Dogaru, NIMB, Volume : 268 Issue : 7-8, (2010) 863-866

## AMS radiocarbon dating of mortar and plaster : the case study of the Modena medieval unesco site.

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[3]Seconda Università degli Studi di Napoli, Dipartimento di Matematica e Fisica, CIRCE lab Caserta, (Italy)

[4]Università degli Studi "Sapienza", Dipartimento di Scienze della Terra, Roma,(Italy)

The carbon dioxide absorbed during the setting of a lime mortar reflects the content of  $^{14}\text{C}$  in the atmosphere at the time of construction of a building. For this reason, the  $^{14}\text{C}$  dating of the mortar is used more and more often in archaeological and architectural research.. The mortars, however, also contain contaminants that could distort radiocarbon dating : fragments of unburnt limestones of geological origin mixed as aggregate may result in older age than expected. On the other hand the re- crystallization and neomorphism of calcite may produce younger dates due to exchanges with the atmosphere after the construction. The Centre for Isotopic Research on Cultural and Environmental heritage of SUN has recently obtained some promising results in the radiocarbon dating thanks to the development of a procedure aiming to eliminate contamination that may occur in a mortar. The construction history of the UNESCO World Heritage Site of Modena (Italy) is still controversial and represents a challenging case study for the application of absolute dating methods for several reasons. The mortars contain a high percentage of carbonate aggregate and thus a proper sample preparation procedure is essential to avoid all the possible contamination risks. AMS Radiocarbon dating carried out on mortar and lumps helped to verify several building phases for the medieval Cathedral and the Ghirlandina Tower in the UNESCO site of Modena. Detailed petrographic characterization of about 150 samples of mortar has allowed us to demonstrate the existence of at least two phases of construction characterized by different production technologies and supply sources, and was used as the basic scientific information for the absolute dating methodologies on mortars and plasters.

Topic : GAA 31

Session 10B

## How old is the human heart ?

Buchholz Bruce.<sup>1</sup>

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Two published bomb-pulse dating studies of cardiomyocyte lifetimes yield significantly different results. Our laboratory performed similar  $^{14}\text{C}$ -AMS analyses of the DNA samples presented by the research groups from the Karolinska Institute and Harvard University. The studies differed in the method employed to isolate the cells of interest, cardiomyocytes, the procedure for isolating DNA for bomb pulse dating, and the mathematical models employed in data interpretation. Since the heart is a relatively large organ, ample DNA was available for  $^{14}\text{C}$ -AMS analyses. The Harvard group isolated cardiomyocytes by cell size while the Karolinska group lysed the cells and used fluorescence activate cell sorting (FACS) to sort on a specific fibrotic protein associated with the surface of cardiomyocyte cell nuclei. The DNA extractions were different but both have long histories of use. Both the phenol-chloroform method (Karolinska) and sodium iodide method (Harvard) are well established. Modeling the  $^{14}\text{C}$  data was significantly different in the two studies. Because  $^{14}\text{C}$  analyses yield a weighted average of the cell population, the measured  $F^{14}\text{C}$  of a DNA sample can be produced by different cell cycling scenarios. The influence of the differences in the studies will be discussed. A short guide for AMS personnel involved in cell turnover studies will be presented.

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Topic : GAA 41

Session 10B

**Bringing AMS radiocarbon into the anthropocene : potential and drawbacks in the determination of the bio-fraction in industrial emissions and in carbon-based products****G. Quarta,<sup>1</sup> G. Ciceri,<sup>2</sup> V. Martinotti,<sup>2</sup> M. D'Elia,<sup>1</sup> L. Calgagnile.<sup>1</sup>**

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[2]RSE (Research of the Energetic System) SpA, Milan,Italy

In the frame of the general efforts to reduce atmospheric CO<sub>2</sub> emissions different efforts are being carried out to stimulate the use of non-fossil energy sources and raw materials. Among these a significant role is played by the use of bio-based fuels and of waste in Waste to Energy plants. In this case a relevant problem is related to the determination of the proportion between the bio and the fossil derived fraction in CO<sub>2</sub> atmospheric emissions since only the share of energy derived from the bio-fraction combustion can be labeled as "renewable". We discuss the potential of radiocarbon in this field by presenting the results of different campaigns carried out by analyzing CO<sub>2</sub> sampled at the stack of different power plants in Italy and bio-fuels with different expected bio-fractions. Despite some drawbacks, such as those related to proper mass fractionation correction and data analysis procedures, this is a fast emerging field of application for <sup>14</sup>C, also considering that it is included among the protocols certified at the international level.

Topic : PRE 05

Session 10B

**Progress report on a novel in-situ  $^{14}\text{C}$  extraction scheme at the University of Cologne.****Fueloep Reka-Hajnal,<sup>1,2</sup> Dunai J. Tibor,<sup>1</sup> Wacker Lukas.<sup>3</sup>**

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[3]Ion Beam Physics, ETH Zürich (Switzerland)

We present initial results of in situ  $^{14}\text{C}$  system blank and calibration sample measurements obtained using the in situ  $^{14}\text{C}$  extraction system developed at the University of Cologne. The  $^{14}\text{C}$  extraction scheme specifically exploits the phase transformation of quartz to cristobalite in order to quantitatively extract the carbon as carbon dioxide and follows a scheme that is different to that of existing extraction systems. Features are offline furnace extraction, single pass catalytic oxidation using mixed copper(I,II) oxide as catalyst, the consequent use of UHV-compatible components and of vacuum annealed copper tubing. The design allows a relative rapid sample throughput - two samples per day as opposed to the current two days per sample - and can accommodate samples ranging between 0.5 to 4 grams of clean quartz. Following extraction and cleaning, the carbon dioxide gas is measured using the gas ion source of the MICADAS AMS facility at ETH Zurich. The extraction system yields low systems blanks (currently averaging around  $4 \times 10^4$  atoms of  $^{14}\text{C}$ ) and the initial results indicate that levels as low as  $1 \times 10^4$  atoms of  $^{14}\text{C}$  are achievable. Measurements of the CRONUS-A standard sample show a good reproducibility and results are consistent with published values. Results of analyses from an old saturated sample collected from a bedrock surface in the Namib desert yield an average  $^{14}\text{C}$  spallogenic production rate of  $\sim 13.9 \pm 2$  atoms  $\text{g}^{-1} \text{yr}^{-1}$ , which is consistent with recently published values.



Topic : SP 02

Session 10B

**A tale of tar : collagen extraction from asphalt-impregnated bones.****Southon John,<sup>1</sup> Fuller Ben,<sup>1</sup> Harris John,<sup>2</sup> Farrell Aisling,<sup>2</sup> Campbell Ken.<sup>3</sup>**

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The Rancho La Brea Tar Pits in the heart of Los Angeles contain one of the largest concentrations of floral and faunal remains from the late Pleistocene. The material was preserved in episodic crude oil seeps that pooled as asphalt deposits at the surface and acted as traps, capturing samples from the entire local ecosystem from leaves and insects through mammoths and mastodons. Although no soft tissues have survived impregnation by asphalt, leaves and wood, insect chitin, and collagen in bird, reptile, and mammal bones are all exceptionally well preserved. However, churning within the deposits has led to mixing of samples of very different ages, and asphalt removal for accurate radiocarbon dating and paleodietary studies using stable isotopes poses a significant challenge. Here we report a novel technique for isolating bone collagen from asphalt contaminated bones that is far simpler and less time consuming than methods used previously, and we discuss some of the initial radiocarbon and stable isotope results.

Topic : SP 04

Session 10B

## Extraction and analysis of sub-milligram per litre concentrations of Methane from Groundwater for $^{14}\text{C}$ analysis.

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Methane ( $\text{CH}_4$ ) is formed biologically and non-biologically in groundwater. While pure  $\text{CH}_4$  end-members have distinct  $\delta^{13}\text{C}$  and  $\delta\text{D}$  isotopic signatures, use of  $\delta^{13}\text{C}$  and  $\delta\text{D}$  alone is not always sufficient to identify the route of formation, as groundwater mixing and/or the presence of methanotrophic bacteria in some systems can confound the isotopic signals. Radiocarbon analysis can be used to help identify sources of methane in groundwater systems as it reflects the age of the source carbon irrespective of route of formation. Interest in this area of research is growing due to the importance of  $\text{CH}_4$  as a greenhouse gas, the increased exploitation of unconventional gas reserves, (e.g. "fracking") and recognition of the extent and importance of the subsurface biosphere.  $\text{CH}_4$  is present in groundwater systems in a wide range of concentrations. Currently, above  $1 \text{ mg C l}^{-1}$  ( $1333 \mu\text{g CH}_4 \text{ l}^{-1}$ ) radiocarbon analysis by Accelerator Mass Spectrometry (AMS) is straightforward. Below this concentration however, radiocarbon analysis becomes more challenging, as large volumes of groundwater are needed to provide sufficient methane for conversion to graphite, background contamination is more significant and the low weight of sample graphite presents challenges during AMS measurement. We describe a new method for field extraction of dissolved  $\text{CH}_4$  for radiocarbon analysis, when it exists at concentrations above  $3 \mu\text{g CH}_4 \text{ l}^{-1}$ . We present  $^{14}\text{C}$  signatures from groundwaters of the Upper and Lower Greensand aquifers in England where concentrations ranged from  $\sim 3$  to  $7 \mu\text{g CH}_4 \text{ l}^{-1}$ , resulting in the production of graphite targets with carbon weights ranging from 0.11 to 0.31 mg C and  $^{14}\text{C}$  values of between  $\sim 2$  and 62 % modern carbon.

## Continuing developments in the $^{14}\text{C}$ community inter-comparisons (SIRI)

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The sixth (SIRI) radiocarbon laboratory inter-comparison extends previous radiocarbon international quality assurance programmes. The main aims and objectives of SIRI are : (1) to demonstrate the comparability of routine analyses carried out in radiocarbon laboratories (2) to quantify the extent and sources of variation in results (3) through choice of material to contribute to the discussion concerning laboratory offsets and error multipliers in the context of IntCal (the International Calibration Programme). (4) to gain a better understanding of differences in background derived from a range of infinite age material types SIRI is a single stage proficiency trial, which started in 2013 with the distribution of samples to the participating laboratories. Samples include a sequence of single tree rings, bones, humic acid and charcoal including several background, and close to background samples. The seven wood samples span Medieval to background ; several are single rings, others decadal. They come from New Zealand, Europe and the United States. The two bone samples are both anticipated to be close to background. The charcoal sample is from a European Palaeolithic site. A doublespar, a humic acid and a barley mash sample make up the set of the thirteen samples which have been distributed to more than 60 laboratories worldwide. The results of the analysis to explore the extent of variation, any laboratory offsets and to quantify the differences in background for the infinite age samples will be presented.

**17.7 Thursday 28 August - Morning****Topic : ISSI 06****Session 11****Anion Formation by Neutral Resonant Ionization.****Vogel John,<sup>1</sup>**

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Resonant transfer of electrons occurs if the total internal energy remains nearly constant in a collision, as for an electron affinity (EA) of one atom that is equal to the ionization potential (IP) of a colliding atom. Middleton & Klein [1], suggest that the blue plasma above recessed samples in Cs sputter ion sources is the location of anion formation, but fail to explain how charge exchange occurred at eV energies. Cs atoms evaporated from a hot sample reach an optically thick density of  $>10^{13}$  Cs per  $\text{cm}^3$  to form a plasma powered by secondary sputter electrons ( $\approx 2$  eV). A collision-radiation model of this plasma was dominated by highly excited Cs with low IP (2). The majority of sputtered sample atoms are low energy ( $<5$  eV) neutrals that are efficiently anionized by the  $\text{Cs}^*$  plasma having IP's similar to EA's of sputtered neutrals. The energy deficit (EA-IP) in carbon-Cs atomic collisions is reduced from 2.6 eV for Cs (6s) to 0.1 - 0.8 eV for the excited  $\text{Cs}^*$  (7d, 8p, 5d, 6p, 7p), predicting a 1500% increase of  $\text{C}^-$  from the plasma over that from a planar sample without plasma. The model mimics the high intensity Cs sputter sources that produce  $>300 \mu\text{A C}^-$ , even down to explaining pulsing ion current and the very high pulse of  $\text{C}^-$  before the beam settles into constant current. Isotopic independence is predicted by the model. An unusual extraction electrode may be effective in propagating this independence.

[1]. R. Middleton &amp; J. Klein, Phys. Rev. A 60 : 3786 (1999).

[2]. J. Vogel, AIP Conf. Proc. 1515 : 89 (2013).

Topic : AAT 01

Session 11

**Ion Source Development for Ultratrace Detection of Uranium and Thorium.**

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Ultrasensitive analytical techniques are required to measure the impurity levels in the high purity Cu materials to be used for the experiment to search for neutrinoless double-beta-decay [1]. We are studying the feasibility of quantitatively determining the U and Th impurities in the Cu by AMS. To achieve ultra low detection limits for U and Th, the use of high efficient positive ion sources is considered. This approach takes advantage of the former HRIBF at ORNL where various positive ion sources have been used to generate ion beams of exotic nuclei. The positive ions can be converted to negative ions by charge exchange and sent to the 25-MV Tandem. The positive-ion sources have the potential of 10-100 times improvement in ionization efficiency for actinides over Cs-sputter negative-ion sources. Two positive-ion sources are being investigated : a hot-cavity surface ionization source and a resonant laser ionization source. In initial studies, we have obtained overall ionization efficiencies of 2-3% for U and Th. Higher efficiency is expected by optimizing the cavity material and geometry as well as the operating conditions. Ionization efficiencies on the order of 1% have been reported for U and Th with a resonant ionization source [2]. More efficient ionization schemes are under development. An important advantage of the laser ion source is its elemental selectivity to suppress the interfering and background ions.

[1] S. R. Elliott, Advances in High Energy Physics, Article ID 365432, 2014. doi :10.1155/2014/365432.

[2] S. Raeder, Ph. D. thesis, 2010, University of Mainz, Germany.

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## From tip to toe – Improvements of the DREAMS facility for the determination of volatile and heavy radionuclides.

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Since the DREAMS (DREsden Accelerator Mass Spectrometry) facility [1] based on a HVE 6 MV Tandetron went operational in 2011, special effort was immediately devoted to upgrading the system for measurements of volatile elements e.g. Cl, I, and heavy elements e.g. actinides. In the case of volatile elements, understanding and minimizing the ion source memory effect is a key issue for precise AMS-measurements [2,3]. For this purpose, one of the two original HVE sources was mechanically optimised. The new design has a more open geometry to improve the vacuum level and a modified target loading and positioning system, which allows exchanging the cathode aperture together with each target. To evaluate improvements of these modifications in comparison to other up-to-date AMS facilities [4], the long-term memory effect in the ion sources of VERA [5], ASTER [3] (Accélérateur pour les Sciences de la Terre, Environnement, Risques) and DREAMS [1] has been investigated by measuring samples of natural  $^{35}\text{Cl}/^{37}\text{Cl}$ -ratio and samples containing highly enriched  $^{35}\text{Cl}$  ( $^{35}\text{Cl}/^{37}\text{Cl} \sim 1000$ ). In these measurements the modified DREAMS ion source showed the lowest level of ion source memory effect and typically the fastest recovery [4]. To extend the measurement capabilities to actinides a time-of-flight system based on thin carbon foils and Micro Channel Plates was designed and constructed at DREAMS. For an optimal tuning of the system with low currents special beam diagnostic elements were manufactured. In cooperation with ANU first actinide samples were measured at DREAMS.

[1] S. Akhmadaliev et al. NIMB 294 (2013) 5.

[2] R. Finkel et al. NIMB 294 (2013) 121.

[3] M. Arnold et al. NIMB 294 (2013) 24.

[4] S. Pavetich et al. NIMB, NIMB 329 (2014) 22.

[5] M. Martschini et al. NIMB 269 (2011) 3188.

## Preparation of carrier free iodine target for speciation analysis of $^{129}\text{I}$ in environmental samples by AMS.

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[3]China Institute for Radiation Protection (China)

Iodine-129 is an important radionuclide in nuclear waste depository, environmental and oceanographic tracer studies. In these studies, speciation analysis of  $^{129}\text{I}$  in soil, sediment and seawater is required. For the determination of ultra-low level  $^{129}\text{I}$  or directly measurement of  $^{129}\text{I}/^{127}\text{I}$  atomic ratio, carrier free iodine has to be separated from the samples, and prepared as a suitable target for AMS measurement of  $^{129}\text{I}$ . A series of methods have been developed in our laboratory for separation of different species of iodine from soil, sediment and seawater without addition of stable  $^{127}\text{I}$  carrier. For soil and sediment, water soluble, exchangeable, carbonate, metal oxides, organic matter and mineral associated iodine were separated by a modified sequential extraction method. The separated iodine in solution was directly precipitate as AgI-AgCl-Ag<sub>2</sub>SO<sub>3</sub> co-precipitate after conversion of extracted iodine to iodide and addition of 0.5-1.0 mg of chloride and NaHSO<sub>3</sub>. For seawater samples, iodide was selectively separated by co-precipitate of AgI with Ag<sub>2</sub>SO<sub>3</sub> and AgCl by addition of only less than 100 mg Ag<sup>+</sup> to up to one liter seawater. The control of the pH value and NaHSO<sub>3</sub> concentration are the critical parameters for effective separation of carrier free iodide from seawater without crossover of iodate into the precipitate. Iodate in the supernatant is separated using the same method after reduction of iodate to iodide using NaHSO<sub>3</sub> in acidic medium. The Ag<sub>2</sub>SO<sub>3</sub> and AgCl in the co-precipitate were washed out using water and ammonium. 1-3 mg of AgI-AgCl precipitate was finally obtained for AMS measurement of  $^{129}\text{I}$ . The developed method has successfully used for the determination of  $^{129}\text{I}$  in soil depth profile and depth profiles of seawater collected in the Antarctic.

Topic : AAT 03

Session 11

# I/Te separation in an RFQ gas cell and the potential use of $^{125}\text{I}$ as a spike for AMS analysis of $^{129}\text{I}$ at low levels.

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$^{125}\text{I}$  ( $T_{1/2} = 59.4$  d) is a readily accessible radioisotope of iodine used in medical imaging, cancer therapy, and as a yield tracer in iodine carrier-free sample preparation techniques for  $^{129}\text{I}$  analysis by AMS. However for measuring low-level samples, it would be further advantageous if AMS could also measure  $^{125}\text{I}$  as an internal reference, to which  $^{129}\text{I}$  (and  $^{127}\text{I}$ ) could be normalized. In this case only a minute amount of  $^{125}\text{I}$  needs to be added to eliminate the  $^{129}\text{I}$  contamination introduced during sample preparation. The direct counting of  $^{125}\text{I}$  ions by AMS, however, requires an on-line isobar separation technique to eliminate the  $^{125}\text{Te}$  interference, a task now made possible with the RFQ gas cell technique currently under development. With this technique, over six orders of magnitude suppression of  $\text{S}^-$  in  $\text{NO}_2$  with respect to  $\text{Cl}^-$  has been demonstrated [NIM B268 (2010) 839]. Because the chemical properties of the  $\text{I}^-/\text{Te}^-$  pair are expected to be similar to that of  $\text{Cl}^-/\text{S}^-$ , we have experimented with the suppression of  $\text{Te}^-$  over  $\text{I}^-$  in  $\text{NO}_2$  and have found, again, five orders of magnitude relative suppression. Furthermore from test samples containing calibrated quantities of  $^{125}\text{I}$ , we have demonstrated the unambiguous measurement of  $^{125}\text{I}$  in proportion to the quantity introduced, and have confirmed that the addition of minute amounts of  $^{125}\text{I}$  is, in fact, free of  $^{129}\text{I}$  introduction. The details of the experimental procedures and instruments will be discussed, as will new geophysical applications that this method may make possible.



Topic : GAA 29

Session 12A

**Induced nuclide  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{22}\text{Na}$  in a granite core exposed by 160 GeV/c muon**

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Horiuchi Kazuho,<sup>4</sup> Matsuzaki Hiroyuki,<sup>5</sup> Sasaki Nobuyoshi,<sup>4</sup> Ohe Ko,<sup>3</sup> Sato Taiichi,<sup>1</sup> Kondo Kaoru,<sup>3</sup>  
Tokanai Fuyuki,<sup>3</sup> Gunji Syuichi,<sup>3</sup> Naoyoshi Iwata,<sup>6</sup> Kazuo Nakashima,<sup>6</sup> Masao Ban,<sup>6</sup> Yasuhisa  
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Cosmogenic nuclide is a powerful tool to investigate secular variation of cosmic rays in the Galaxy over millions of years. In particular, since high energy cosmic rays above 1TeV is insensitive for the solar modulation because of the Larmor radius beyond the size of heliosphere, their secular variations provide us information about the origin and propagation in the scale of galaxy. High energy muons above 100 GeV/c produce nuclide such as  $^{10}\text{Be}$  and  $^{26}\text{Al}$  through interactions with  $\text{SiO}_2$  in rocks being at deep underground. Since the half-life of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  are  $1.6 \times 10^6$  and  $7.2 \times 10^5$  years, respectively, using the traces printed in rocks by muon exposure, we can find out secular variations of high energy cosmic rays over the past millions of years. For the study, it is important to know production process of nuclide in granite by muon exposure. We have experimentally investigated the production rates of nuclide  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{22}\text{Na}$ , exposing the 160 GeV/c muon beam to an 1 m long granite core at the COMPASS experiment beam line in CERN. Approximately, muons were irradiated to the core over 100 days. The  $^{22}\text{Na}$  with short half-life was measured using high purity Germanium-gamma ray detector installed in a low background shielding system. The  $^{10}\text{Be}$  and  $^{26}\text{Al}$  were analyzed using Accelerator Mass Spectrometry in MALT, Tokyo University (AMS) after a chemical treatment for the extracted  $\text{SiO}_2$  from the core. The production rates per muon were  $(2.41 \pm 0.09) \times 10^{-8}$  atoms/g,  $(0.75 \pm 0.011) \times 10^{-8}$  atoms/g,  $(5.4 \pm 0.11) \times 10^{-8}$  atoms/g for  $^{22}\text{Na}$ ,  $^{10}\text{Be}$ , and  $^{26}\text{Al}$ , respectively, in synthetic silica. We describe the production rates of nuclide in the granite core.

Topic : GAA 18

Session 12A

**Production rate of  $^{10}\text{Be}$  in magnetite.****Granger Darryl,<sup>1,2</sup> Riebe Clifford,<sup>3</sup> Moore Angus,<sup>1</sup> Rogers Heather,<sup>3</sup> Lifton Nathaniel.<sup>1,2</sup>**

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Beryllium-10 is commonly measured in quartz for determining exposure ages and erosion rates, but this limits its use to quartz-bearing rocks. Magnetite ( $\text{Fe}_3\text{O}_4$ ) is a common mineral that is resistant to weathering, contains abundant oxygen as a target for  $^{10}\text{Be}$  production, and is relatively easily separated from other minerals. Importantly, magnetite can be found in many rocks where quartz is absent, thus measurement of  $^{10}\text{Be}$  could open up new opportunities for measuring erosion rates in landscapes that have previously been inaccessible or difficult for cosmogenic nuclide methods, including landscapes on volcanic and ultramafic rocks. Production rates were determined for a granitic boulder from Mt. Evans, Colorado, USA, previously collected by D. Elmore and D. Lal. We separated magnetite from finely pulverized rock using a combination of hand magnets and selective chemical dissolution in dithionite-citrate-bicarbonate solution, 5% nitric acid and 1% hydrofluoric and nitric acid. Three aliquots that did not receive HF treatment were contaminated with meteoric  $^{10}\text{Be}$ , most likely contained in small amounts of mica. Three aliquots that received HF treatment agreed to within 2% measurement error. The relative production rate by mass of  $^{10}\text{Be}$  in magnetite and quartz at Mt. Evans is  $0.462 \pm 0.012$ . Our results are somewhat lower than theoretically predicted values based on excitation functions for O, Si, and Fe, suggesting that production by neutron spallation on Fe is probably overestimated. Additional samples of magnetite and quartz from both bedrock and sediment are in preparation.

Topic : GAA 38

Session 12A

 **$^{10}\text{Be}$  in polar ice as proxy or solar activity.****Heikkilä Ulla,<sup>1</sup> Smith Andrew,<sup>1</sup>**

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Meteoric  $^{10}\text{Be}$  is a commonly used proxy for solar activity. The records, stored in natural archives such as ice cores, reflect solar cycles but some noise is added to the signal due to the transport through the atmosphere and deposition into the ice. In this study we address this noise by means of atmospheric transport modelling, comparison of records from various parts of the world as well as comparison of  $^{10}\text{Be}$  with another cosmogenic radionuclide,  $^{14}\text{C}$ . We inspect different time periods ranging from seasonal and annual to the Holocene and beyond, which reveals solar cycles from the 11-year one to the longer (hundred to thousands of years) ones. The results show that the deposition from the atmosphere into the ice is not largely influenced by climatic factors, however the actual snow concentrations can be biased by significant changes in snow accumulation, for example during glacial-interglacial transitions.

Topic : GAA 12

Session 12A

## Solar activity over the last millennium based on a new $^{10}\text{Be}$ record from Dome C (Antarctica).

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[@] ASTER Team : M. Arnold, G. Aumaître, D.L. Boulès, K. Keddouch.

We will present a new  $^{10}\text{Be}$  record covering the last millenium from an ice core from Dome C (Antarctica). The four minima of solar activity (Wolf, Spörer, Maunder and Dalton) known over the studied time period are evidenced by an increase of 7% of the  $^{10}\text{Be}$  concentration compared to the average concentration, in agreement with previous studies at South Pole and Dome Fuji in Antarctica (Bard et al., 1997; Horiuchi et al., 2008) and at NGRIP and Dye3 in Greenland (Berggren et al., 2009). The annual to biennial resolution allows in addition detecting the 11-year solar cycle. Sulfate concentration measured in the exact same samples enables a direct comparison of both  $^{10}\text{Be}$  and sulfate profiles which corroborates the systematic relationship between stratospheric eruptions and  $^{10}\text{Be}$  concentration increases as recently highlighted by Baroni et al. (2011) regarding the stratospheric volcanic eruptions of Agung in 1963 and Pinatubo in 1991. This is probably due to an increase of the  $^{10}\text{Be}$  deposition flux related to a significant volcanic aerosols sedimentation enhancement. The presented new record thus confirms the need to identify factors other than solar that may influence the  $^{10}\text{Be}$  signal before using it as a proxy for the solar activity and irradiance.

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Topic : GAA 21

Session 12A

**Long-term waterfall dynamics in monsoonal Australia based on cosmogenic  $^{10}\text{Be}$ .****Fujioka Toshiyuki,<sup>1</sup> May Jan-Hendrik,<sup>2</sup> Fink David,<sup>1</sup> Nanson Gerald,<sup>2</sup> Jansen John,<sup>2</sup> Codilean Alexandru.<sup>2</sup>**

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Extensive plateaus, arrays of escarpments and a variety of waterfalls are iconic to northern Australia. How old and stable are these features? Tectonically, northern Australia has been quiescent during the Quaternary. Rainfall is highly seasonal and dominated by the summer monsoon. In this setting, regional landscape dynamics should be strongly affected by fluctuations in monsoon and the associated fluvial processes. Here, we examine timescales and processes of waterfall evolution in northern Australia. Situated in the Kimberley sandstone plateau, Durack Falls comprise a series of 1-3 m falls, while Bindoola Fall is a large ~15 m fall. Surprise Creek, ~100 km south of Darwin, has three 3-5 m waterfalls with deep plunge pools developed at the edge of a quartzite plateau. Over 30 samples were collected from bedrock straths up- and downstream of the waterfalls and on their headwall. Their  $^{10}\text{Be}$  exposure ages (assuming zero erosion) reveal contrasting results. While two waterfalls in the Kimberley show relatively young, variable ages (15-110 ka for Durack and 11-57 ka for Bindoola), Surprise Creek indicates old, but uniform ages (94-160 ka). Out-of-channel, undisturbed bedrock exhibits consistently high  $^{10}\text{Be}$  equivalent to steady-state erosion rates of 2-5 mm/ka, in agreement with typical bedrock erosion rates observed across Australia. Based on these data, we here present a model to evaluate process and rates of waterfalls formation, and discuss the controlling factors.

Topic : PRE 03

Session 12B

**Status of the 3MV multi-element AMS in Xi'an, China.**

**Lu Xuefeng<sup>1,2</sup> Wu Zhenkun<sup>1,2</sup> Liu Qi<sup>1,2</sup> Fu Yunchong<sup>1,2</sup> Zhao Wennian<sup>1,3</sup> Huang Chunhai<sup>1,3</sup> Zhao Xiaolei<sup>1,4</sup> Zhou Weijian.<sup>1,2</sup>**

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[4]IsoTrace Laboratory, University of Toronto (Canada)

The Xi'an 3MV AMS facility has been in operation since 2006. It is the 4th AMS system designed by High Voltage Engineering Europa (HVEE) that includes a sequential-injection system (i.e. a "bouncer injection"). The facility is a multi-element system with a single beam line dedicated to  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$  and  $^{129}\text{I}$  analysis. Thus far, there are about 19,000  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$  and  $^{129}\text{I}$  targets have been measured. We report here on the status and performance of the facility, troubles, technical improvements and a precision study on modern environment samples.

Topic : PRE 06

Session 12B

### The first three years of CologneAMS.

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The CologneAMS laboratory is operational since mid 2011. The system was designed for a wide range of applications. In our first three years we did routine measurements of several isotopes which are Be, C, Al and Pu. This list will be extended in the future. The quality of the routine measurement with respect to blank values and reproducibility is presented.

A TOF-BPM-System was developed as a general tool for future measurements. We used this TOF-system for high quality measurement of energy loss and straggling in different materials. Results for different isotopes will be compared to calculated values using different approaches. With this data at hand we plan to optimize the setup for isobar suppression for different isotopes.

We give an overview of our activities concerning benchmarks of the quality of the measurement as well as different developments of our experimental setups.

### Status report of the 1 MV AMS facility at CNA.

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[4]AMS <sup>14</sup>C Dating Centre, Department of Physics and Astronomy, Aarhus University, (Denmark).

SARA (Spanish Accelerator for Radionuclide Analysis) was the first multielemental AMS facility installed in Spain in 2005. Since then it has been dedicated to the routine analysis of several radionuclides, such as <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>129</sup>I and Pu isotopes [Chamizo et al. 2008]. Tests have been carried out with other isotopes, such as <sup>41</sup>Ca, <sup>236</sup>U and <sup>237</sup>Np. Several changes have been made to the original facility to improve performance. First, an upgraded version of the ion source SO-110 has allowed us more stable measurement conditions for volatile elements, and a better general performance. Besides, changes in the target geometry have improved the ionisation efficiency and long-term stability of the source output. Moreover, different software upgrades have been introduced to meet our routine operational needs. Finally, changing the movable Faraday-cup associated electronics now allows the measurement of smaller stable isotope currents (in the range of the pA), which has been key for the study of <sup>236</sup>U/<sup>238</sup>U atomic ratio in environmental samples. Apart from these modifications it has to be noted that routine radiocarbon measurements have been moved to a Micadas system (200 kV) installed at CNA in 2012. In this paper we will illustrate the evolution of the facility up to now, and our future prospects will be introduced.

References : E. Chamizo. J.M. Lopez-Gutierrez. A. Ruiz-Gomez, F.J. Santos, M. Garcia-Leon, C. Maden, V. Alfimov. (2008) Status of the compact 1 MV AMS facility at the Centro Nacional de Aceleradores (Spain). Nucl. Instr. and Meth. B266 2217-2220.



Topic : PRE 01

Session 12B

**Equipment upgrades at the UC Irvine Keck AMS laboratory.****Southon John,<sup>1</sup> Santos Guaciara,<sup>1</sup> McCormick Cyril,<sup>1</sup> Pederson Chris,<sup>2</sup> Roberts Mark.<sup>3</sup>**

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The Keck AMS laboratory at the University of California Irvine operates an NEC 0.5 MV Compact AMS system optimized for high precision high throughput  $^{14}\text{C}$  measurements. We have recently carried out several upgrades to increase sample throughput, reduce down time, and improve ease of spectrometer tuning. These include installation of a gas detector, a home-built 60 sample version of the NEC MC-SNICS ion source, and a large diameter gas stripper with two stages of differential pumping. These upgrades will be discussed along with other system improvements and lessons learned from some unusual equipment failures.

Topic : PRE 02

Session 12B

**Performance of the rebuilt SUERC single-stage accelerator mass spectrometer.**

Shanks Richard,<sup>1</sup> Ascough Philippa,<sup>1</sup> Dougans Andrew,<sup>1</sup> Gallacher Paul,<sup>1</sup> Gulliver Pauline,<sup>1</sup> Rood Dylan,<sup>1</sup> Xu Sheng,<sup>1</sup> Freeman Stewart.<sup>1</sup>

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The SUERC bipolar single-stage accelerator mass spectrometer (SSAMS) has dismantled and rebuilt to accommodate an additional rotatable low energy electrostatic analyser. This is to suppress oxygen interference to radiocarbon measurement and facilitate the attachment of a developmental positive ion source in addition to a Cs<sup>-</sup>sputter source. The spectrometer up-grade and performance will be discussed along with additional alternative applications.

Topic : MNSI 01

### Radiocarbon measurements at LAC-UFF : recent performance

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[4]Escola Superior de Agricultura Luiz de Queiroz, Departamento de Ciências Florestais, Universidade de São Paulo (ESALQ-USP),( Brazil)

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In 2012 a NEC 250KV SSAMS system was installed at IF-UFF, Niteroi, Brazil. After installation, typical processing blanks were  $1.3 \times 10^{-14}$  while reference materials indicate precision and accuracy of just 0.8% [1]. Here we report tests performed both on the combustion protocol and on the machine parameters at LAC-UFF. The main goal was to reduce the background when processing organic samples, and to improve spectrometer accuracy and precision. To minimize the blank, we conducted investigations on the combustion step. By baking Ag wire and CuO separately prior to loading with samples and sealed under vacuum, we attained a background of  $8 \times 10^{-15}$ , when processing  $^{14}\text{C}$ -free organic samples. To investigate spectrometer accuracy and precision, we measured a selected set of annual tree-rings between 1927 and 1997 of *Araucaria Angustifolia*, a tree species from Southern of Brazil [2]. This set of sample represents a good benchmark to evaluate the performance of the machine since high precision and accuracy are required for reliable dates. A quite good agreement is observed between LAC-UFF and KCCAMS datasets although online  $\delta^{13}\text{C}$  values seem to be a major limitation for accuracy at LAC-UFF. Further tests were performed aiming to understand and minimize the machine fractionation effect. Typical SSAMS measurements have been carried out at 20-25mA  $^{12}\text{C}^{+1}$ , but such currents may systematically affect the  $^{13}\text{C}/^{12}\text{C}$  ratios [3]. Measurements were performed at lower currents, 7-15mA  $^{12}\text{C}^{+1}$ , but no systematic change for  $^{13}\text{C}/^{12}\text{C}$  ratios has been observed in our machine.

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**17.8 Thursday 28 August - Afternoon****Topic : GAA 79****Poster Session 2****Fish otoliths as radiocarbon referential age markers and palaeothermometers : the Mainitiba I shellmound, in the Southeastern coast of Brazil**

**Aguilera Orangel,<sup>1</sup> Carvalho Carla<sup>2,3</sup> Macario Kita<sup>3,4</sup> Ghosh Prosenjit,<sup>5</sup> Marques Jr Aguinaldo,<sup>1</sup> Souza Rosa,<sup>1</sup> Chanca Ingrid,<sup>3</sup> Monteiro Cassiano,<sup>1</sup> Silva Edson.<sup>1</sup>**

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The Brazilian coast was occupied in the Holocene by fishermen and mollusk gatherers who used to build shell-mounds from food remains. These archaeological sites, found today all over the southern and southeastern coast, are a unique context where well preserved shells and fish otolith can be studied for its biodiversity and deposition characteristics. In this work otoliths were analyzed to evaluate the potential use for radiocarbon referential age markers and palaeothermometers. The Manitiba I shellmound is located by the Saquarema lagoon, Rio de Janeiro state, Brazil, and was previously studied by Kneip (2001) so the specimens analyzed from this site came from the zooarchaeological collection of the National Museum of the Rio de Janeiro Federal University. Three otolith samples from each archaeological layer were dated at the Radiocarbon Laboratory of the Fluminense Federal University (LAC-UFF). Otolith diphractograms showed almost exclusively Aragonite and trace of Calcite. Graphitized samples were measured in a 250kV Single Stage Accelerator. The results show no dependence with depth, indicating statistical fluctuations are larger than the actual occupational period. Calibrated results range from 4200 to 3600 Cal BP. Stratigraphy presents sterile sand layers between archaeological layers, therefore a sequence model was used for modeling in the OxCal software using the marine13 curve. Average water temperatures were estimated based on d18O in otolith (Ghosh et al. 2007).

Ghosh, P. Eiler, J, Campana, S.E. Feeney, R.F. 2007. GCA 71, 2736-2744.

Kneip, L.M. 2001. O sambaqui de Manitiba I e outros sambaquis de Saquarema, RJ. Serie Arqueologia nº 5, Museu Nacional, UFRJ. 5, 91 p.

Topic : GAA 80

Poster Session 2

**Radiocarbon dating of an ancient tomb in hepu county, China****Ruan Xiangdong,<sup>1</sup> Xiong Zhaoming,<sup>2</sup> Sasa Kimikazu,<sup>3</sup> Shen Hongtao,<sup>4</sup> Guan Yongjing.<sup>1</sup>**

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An ancient tomb belonging to the Han Dynasty was excavated in the damper ridge, Hepu County. Damper ridge is an important archaeological site in Hepu County, Beihai City, in south China's Guangxi Zhuang Autonomous Region. It is believed that Hepu County was the oldest departure point on the ancient maritime trading route during the Han Dynasty (206 BC to AD 220) due to the ideal natural geographical conditions and the existence of a large number of Han tombs. Radiocarbon measurements on shell and wood samples from the Damper ridge site were performed at the Paleo Labo Co. Ltd. Japan, and the Xi'an AMS Center, China. The calendar ages of the samples were determined to be a period from 47 BC to 90AD (95% confidence level) calibrated with Marine13. The results of these measurements are presented and the related chronology is discussed.

Topic : GAA 81

Poster Session 2

### Increase of radiocarbon concentration in tree rings from the Kujawy (SE Poland) around AD 774-775

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Evidence of a rapid increase in atmospheric radiocarbon content in AD 774 -775 was presented by Miyake et al. (2012). An increase of about 12‰ in the  $^{14}\text{C}$  content was observed in annual tree rings from Japanese cedar. Usoskin et al. (2013) report a similar  $^{14}\text{C}$  spike in German oak, and attribute it to exceptional solar activity. If this phenomenon is global in character, such rapid changes in  $^{14}\text{C}$  concentration should be included in the calibration curve. Single-year samples of dendro-chronologically dated tree rings (*Quercus robur*) from Kujawy, a village near Krakow (SE Poland), spanning the years AD 765-796 were collected and  $^{14}\text{C}$  content was measured using the AMS system in the Leibniz Laboratory.

Miyaki F, Nagaya K, Masuda K, Nakamura T. 2012. A signature of cosmic-ray increase in AD 774-775 from tree rings in Japan. *Nature* 486. Pp. 240-242.

Usoskin IG, Kromer B, Ludlow F, Beer J, Friedrich M, Kovaltsov GA, Solanki SK, Wacker L. 2013. The AD775 cosmic event revisited : the Sun is to blame. *Astronomy & Astrophysics* 552 : L3

## Study on monitoring of volcanic activity using $^{129}\text{I} / ^{127}\text{I}$ ratios in crater lake and hot spring at Zao volcano, Japan

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Volcanic activity has become higher at Zao volcano, Miyagi Prefecture, Japan, since January 2013 after the 2011 Tohoku Earthquake. Basic water quality of crater lake and hot spring at Zao volcano have been studied by Tohoku University since the water quality of crater lake are correlating with volcanic activity. As a part of this project, we are trying to monitor the volcanic activity using  $^{129}\text{I} / ^{127}\text{I}$  ratios in crater lake and hot spring of Zao volcano.  $^{129}\text{I} / ^{127}\text{I}$  ratios of hydrothermal at Zao volcano are considered to become lower by the supply of chronologically-old iodine in terms of global iodine cycle. In September 2013, water samples (2 L) were collected from the surface of crater lake (Okama) and Kamoshika Hot Spring in the eastern side of Zao volcano.  $^{129}\text{I} / ^{127}\text{I}$  ratios of Okama and Kamoshika Hot Spring were respectively, estimated to be  $(1.5 \pm 0.4) \times 10^{-9}$  and  $(0.78 \pm 0.15) \times 10^{-9}$ , 500 - 1,000 times higher than the steady-state ratio of sea water ( $1.5 \times 10^{-12}$ ). Since  $^{129}\text{I} / ^{127}\text{I}$  ratio of anthropogenic metric water were over  $9.0 \times 10^{-12}$ , Okama and Kamoshika Hot Spring were very likely to be strong affected by the meteoric water including anthropogenic  $^{129}\text{I}$ . For the monitoring of volcanic activity using  $^{129}\text{I} / ^{127}\text{I}$  ratio, it is necessary to decide the site as few anthropogenic  $^{129}\text{I}$  as possible through the measuring of  $^{129}\text{I} / ^{127}\text{I}$  ratio of the Okama bottom water and some hot spring around Zao volcano. Continuous water quality survey of 1 time for Okama and 1 time per 2 months for hot springs are planned for this year.

## Measurements of cross sections for production of light nuclides by 120 GeV and 400 MeV proton bombardment of Y

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The accumulation of long-lived cosmogenic nuclides, such as  $^{10}\text{Be}$  and  $^{26}\text{Al}$  produced by relatively high energy solar and galactic particles, in terrestrial and extraterrestrial materials enables the investigation of their irradiation histories. Reconstructing the conditions under which cosmogenic nuclides are produced requires production cross sections for each pathway leading to the production of a specific cosmogenic nuclide. These data also have a very practical benefit for health and safety in radiation protection; they serve as a comprehensive nuclear database that can be used to estimate residual radioactivities in accelerator facilities. Additionally, cross sections are indispensable for studying the specific formation mechanisms of these nuclides, where spallation, fission, or fragmentation is a dominant process. The fragmentation process is usually studied by production cross sections of light nuclides which are best measured by AMS. For energies  $>100$  MeV few measurements have been made and published. Models for the production of light nuclide by the fragmentation process can be assessed in the energy range going from  $> 100$  MeV to over 100 GeV. We have measured and report the first  $^{10}\text{Be}$  and  $^{26}\text{Al}$  production cross sections from Y produced by 120 GeV and 400 MeV protons. The proton irradiation at 120 GeV and 400 MeV were performed at Fermi National Accelerator Laboratory (FNAL) and the Research Center for Nuclear Physics (RCNP), Osaka University, respectively. The AMS measurements were performed at MALT (University of Tokyo). We will discuss the production mechanism of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  by spallation and fragmentation in two different kinds of high-energy nuclear reactions, whose energy gap is over two orders of magnitude.



## Annual growth rings in a sample of Paraná pine (*Araucaria Angustifolia*) : towards improving the $^{14}\text{C}$ calibration curve for the Southern Hemisphere

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Present calibration of the  $^{14}\text{C}$  time-scale for the Southern Hemisphere (SH) combines  $^{14}\text{C}$  and dendrochronology analyses from decadal wood samples of 0-1000 cal yr BP. Beyond this dataset, the SH curve was initially expanded back to 11ka cal yr BP based on the Northern Hemisphere dataset and a random effects model (SHCal04)[1]. Recently, the SH curve has been extended to 50ka cal yr BP, with the addition of new tree-ring/ $^{14}\text{C}$  values (SHCal13 curve)[2], and assuming interhemispheric offset similar to those measured for the past 0-2000 cal BP. Nevertheless, a South American  $^{14}\text{C}$  curve from dendrochronologically dated wood is still lacking, especially within the tropical or subtropical zones which should experience seasonal shifts of atmospheric  $\text{CO}_2$ . However, the first step towards improving the calibration of the SHCal curve is to assess the annual makeup of the growth rings of long-lived tree species. This can be achieved by  $^{14}\text{C}$  bomb-pulse dating of individual selected dendrochronologically dated rings. Here we report the first set of high-precision (0.2-0.3%)  $^{14}\text{C}$ -AMS of a single tree growing at 22°50'S, 46°04'W (Camamducaia, Brazil) from 1927-1997. Our  $^{14}\text{C}$  results showed the rise and rapid decrease of atmospheric  $^{14}\text{C}$  associated with the detonations of nuclear weapons during the late 50's, and its subsequent uptake by other large C sinks. The agreement between this record and the SH compilation  $^{14}\text{C}$  dataset shows the potential of this tree species for older chronologies. Presently, this  $^{14}\text{C}$  dataset can be used for the study of the global carbon cycle at this latitude, and for the determination of the growth rate of tropical trees without annual ring patterns.

[1] McCormac et al. 2004 Radiocarbon 46 :1087

[2] Hogg et al 2013 Radiocarbon 55 :1889

## Testing the removal of exogenous bounded carbon from modern human hair by cross-flow nanofiltrated amino acids procedure

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In forensic investigation, when the deceased date of a victim is required, radiocarbon ( $^{14}\text{C}$ ) measurements on modern human tissues such as nails and hair can help determine the year-of-death (YOD) [1]. However, rear-hair which is frequently subjected to cosmetic products that contain petrochemical derivatives as well as plant and animals extracts [2,3], can bias the  $^{14}\text{C}$  results towards depleted values [4]. Currently, the various chemical pretreatments available in the literature are ineffective in removing foreign C contaminants. Exogenous impurities strongly embedded themselves into the hair structure, percolating beyond the cuticle layer [2,3]. Here, we applied cross-flow nanofiltrated amino acid (CFNAA) extractions [5] to keratinaceous tissues from a single human subject, including rear-hair samples contaminated by a permanent coloring from a dark-brown dye kit (rear-hair from subject B, in [4]). In order to investigate if significant discrepancies between contaminated and non-contaminated keratinaceous tissues can be resolved, we conducted isotopic analysis ( $^{14}\text{C}$ ,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and C/N) of solvent treated and CFNAA extracted samples (fingernails, body- and rear-hair). This comparison allow us to determine the efficiency of the CFNAA isolation method when dealing with the removal of petroleum base derivatives from rear-hair, as well as the possibility of using other keratinaceous tissues (fingernails and body-hair) for YOD determinations. These results will be shown and discussed.

[1] Hodgins 2009. NIJ Final Rep.

[2] Kuzuhara and Hori 2003. J. App. Poly. Sci. 90 :3806

[3] Chen et al. 2006. App. Surf. Sci. 252 :6786

[4] Martinez De La Torre et al 2014. Radiocarbon 56 : 53

[5] Boudin et al. 2013. Rapid Commun. Mass Spectrom. 27 : 2039

## The next chapter of direct phytolith $^{14}\text{C}$ dating : debunking the myth of occluded photosynthetic carbon exclusivity

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Radiocarbon dating of carbon (C) encapsulated in phytoliths (phytC) is currently used in many Earth Science disciplines for absolute chronologies and paleoclimatic reconstructions; however, the usefulness of phytC has been hampered by inadequate extraction methods[1] and uncertainties regarding its origin as purely photosynthetic[2,3,4]. An early investigation measuring isotopes from Gramineae spp. grown in free-air C enrichment experiments (FACE), showed that part of its phytC is from a non-photosynthetic source, thus indicating a dual origin [5]. To demonstrate that non-photosynthetic sources within phytC could be from soil C stocks, we measured  $^{14}\text{C}$ -AMS phytC extracted from a set of Sorghum bicolor growing on known  $^{14}\text{C}$  and  $\delta^{13}\text{C}$  bulk substrates and hydroponic solutions. The phytolith concentrates and a silica blank were extracted at UCI, CEREGE and Wisconsin using an improved protocol[1,2]. We also measured  $\text{CO}_2$  fluxes and isotopic signatures of microbial respiration, percentage of biomass and phytolith extracts produced, and isotopic signatures of the local air and bulk-plant during the growing season of 2012. This allowed comparison of the belowground substrate and nutrient C contributions to phytC  $^{14}\text{C}$  results. Meanwhile, NanoSIMS analyses of phytolith polished sections was used to locate phytC in the phytolith siliceous structure. These results will be shown and discussed.

[1] Corbineau et al. 2013 R. Paleobot. Palyn. 197 : 179

[2] Santos et al. 2010 T. Radiocarbon 52 :113

[3] Santos et al. 2012a Biogeosci. 9 :1873

[4] Santos et al. 2012b Biogeosci. Discussion 9 :C6114

[5] Reyerson et al. 2013 AGU Fall meeting 2013 (Abstract ID : 1803125).

Topic : GAA 87

Poster Session 2

## Insecticide Transfer Efficiency and Lethal Load in Argentine Ants

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We characterized trophallaxis between individual worker ants and examined the toxicant load in dead and live Argentine ants in a colonies exposed to two insecticides having different toxicity mechanisms. About 50% of meals with trace levels of <sup>14</sup>C-sucrose, <sup>14</sup>C-hydramethylnon, and <sup>14</sup>C-fipronil were shared between single donor and recipient ants. Dead workers and queens contained significantly more hydramethylnon (122.7 and 22.4 amol/ $\mu$ g ant ; respectively) than did live workers and queens (96.3 and 10.4 amol/ $\mu$ g ant ; respectively), with the highest amounts in the abdomen. Dead workers had significantly more fipronil (420.3 amol/ $\mu$ g ant) than did live workers (208.5 amol/ $\mu$ g ant), but dead and live queens had equal fipronil levels (59.5 amol/ $\mu$ g ant versus 54.3 amol/ $\mu$ g ant ), with the highest amounts of fipronil in the thorax of dead queens and in the head of live queens. Resurgence of polygynous ant colonies treated with hydramethylnon baits may be explained by queen survival of sublethal doses resulting from the slowing of trophallaxis throughout a colony. Bait strategies and dose levels for controlling insect pests can be based on specific toxicant behavior and trophic strategies of the entire colony.

Work was performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

## Online coupling of thermal fractionation - $^{14}\text{C}$ AMS for source apportionment of carbonaceous aerosols

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Carbonaceous aerosols are a fraction of air particulate matter (PM) and can have impact on climate and ecosystems due to their influence on the radiation balance of the earth. They consist of organic carbon (OC) and elemental carbon (EC). Due to their optical properties, they can lead to heating or cooling effect in the atmosphere thus becoming of increasing interest in climate research. Radiocarbon is a long-lived radionuclide that is used for environmental dating. It is also a powerful analytical tool for the detection of fossil materials as in these the pre-existing  $^{14}\text{C}$  has decayed. The evaluation of positive and negative artefacts during OC and EC separation is performed with a thermo-optical OC/EC analyzer (Sunset Laboratory) that produces gaseous  $\text{CO}_2$ . Gaseous  $^{14}\text{CO}_2$  AMS measurements of air samples involve several intermediate steps that aim in the separation and purification of large  $\text{CO}_2$  fractions. We focus on the development of an online automated hyphenation to determine  $^{14}\text{C}$  in the different fractions of carbonaceous PM. The online  $^{14}\text{C}$  analysis of carbonaceous PM requires the development of techniques for the measurement of microgram samples with accelerator mass spectrometry (AMS) with high throughput. In this context, the development of a gas inlet system for direct injection of gases ( $\text{CO}_2$ ) released from the OC/EC analyzer is in progress. In this work, we present details of the current offline source apportionment methodology, which allows the investigation of carbonaceous aerosols from ambient air. We also describe the online, trap - free, approach that will make us benefit by the real time  $^{14}\text{C}$  analysis of the thermograms provided by the Sunset OC/EC analyzer avoiding the hypotheses and laborious work of offline sample preparation.

## Coupling of an elemental analyser with AMS for fast radiocarbon analysis of aerosol samples

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For environmental and climate sciences, it is important to apportion the source of the atmospheric aerosols between wood burning, biogenic emissions and fossil fuel combustion. This can be achieved by analysing radiocarbon in the aerosol by accelerator mass spectrometry (AMS). However, sample preparation is highly effort and time consuming (1 hr/sample of experimental work). Previous works have coupled an elemental analyser (EA) with AMS using a Gas injecting System (GIS). Here, we implemented such technique for the analysis of carbonaceous aerosol samples. Constant and cross contamination models were applied in a single equation to make measurement corrections. The GIS traps the CO<sub>2</sub>, delivered at high flow in helium from the EA, with a zeolite column. Next, the CO<sub>2</sub> is released at high temperature and it expands into a syringe. Helium is added to make a pressurized mixture of 5% CO<sub>2</sub>. Afterwards, the syringe slowly (~40 µL/min) delivers the CO<sub>2</sub> into the gas ion source of an AMS. The EA-GIS-AMS system is fully automatic and requires 10 min/sample. Samples were punched out from quartz filters and wrapped with a tin foil for flash combustion inside the EA. As a first approximation, the constant contamination parameters of the EA-GIS system were found by nonlinear regression of the measured ratio (Rm) vs carbon mass of sodium acetate (blank). After that, cross contamination was introduced by intercalating measurements of blanks and standards (oxa2, C7). Finally, the full model was applied to the data of Rm vs mass. In the case of real samples, the pMC values measured with the EA-GIS method showed a 0.99 :1 relationship with the values measured with our conventional method; therefore, we can apply this faster method for routine analysis without throughput loss.

**$^{36}\text{Cl}$  in deep crustal fluid in Japan : implications for fluid origins**

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The Japanese islands are situated in the northwestern margin of the Pacific Ocean, part of the circum-Pacific orogenic belt, where several plates converge to form a tectonically active region. Such a tectonic setting of Japan makes it especially important to assess the long-term stability of deep geological environment as part of the site characterization for potential nuclear waste repositories. Deep saline groundwaters are widely distributed beneath the Japanese island, which can affect the conditions around repositories. Particularly in the southwest Japan arc, saline deep-seated fluid upwells along tectonic lines and associated faults, mixing into deep groundwater systems in the vicinity. To investigate the source and age of the upwelling fluid, deep groundwaters around major tectonic lines were analyzed for  $^{36}\text{Cl}/\text{Cl}$  ratios. The  $^{36}\text{Cl}/\text{Cl}$  ratios in these groundwaters were mostly in the range between  $\sim 1 \times 10^{-15}$  and  $\sim 1 \times 10^{-14}$ . Chemical and isotopic indices including  $\text{Li}/\text{Cl}$ ,  $\text{Br}/\text{Cl}$ ,  $\delta\text{D}$ - $\delta^{18}\text{O}$  and  $^3\text{He}/^4\text{He}$  of groundwaters were used to constrain the  $^{36}\text{Cl}/\text{Cl}$  ratio of the fluid end member. Overall, the estimated  $^{36}\text{Cl}/\text{Cl}$  ratio of deep-seated fluid is likely to be very low ( $\sim 1 \times 10^{-15}$ ), almost equal to the seawater value, while it varies across locations. Variations in  $^{36}\text{Cl}/\text{Cl}$  ratios may indicate the possible fluid source, upwelling path, and residence time in the crust. The remarkably low  $^{36}\text{Cl}/\text{Cl}$  ratio may imply an association of deep-seated fluid with the mantle.

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Topic : GAA 91

Poster Session 2

### **$^{36}\text{Cl}$ -based ages of seawater component in deep groundwater : examples from coastal sedimentary basins in Japan**

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Climate-driven sea-level fluctuations can have impacts on groundwater flow regimes, especially in coastal areas. A sea level decline leads to a seaward movement of the discharge area of regional groundwater flow system, accompanying increased hydraulic heads in aquifers that enhances deeper groundwater flow. It also brings drastic changes in shorelines and associated topography of coastal areas, which potentially affect groundwater flow regimes. Since coastal areas have a potential to be a candidate site for geological disposal of radioactive waste in Japan, an assessment of the influence of sea-level change on groundwater system is especially important. This study utilizes  $^{36}\text{Cl}$  in coastal groundwater to investigate the past influence of sea-level changes on groundwater systems. Deep groundwaters were collected from typical coastal sedimentary basins in Japan and analyzed for  $^{36}\text{Cl}/\text{Cl}$  ratios. The ages of seawater end member were estimated based on the secular equilibrium  $^{36}\text{Cl}/\text{Cl}$  values calculated from rock composition data for each area. The  $^{36}\text{Cl}$ -based ages calculated for coastal sedimentary basins are generally very old (over several hundreds of kyr), except for the groundwaters obtained in the vicinity of the coast (several tens of kyr). This is contrasted by the predominantly young  $^{36}\text{Cl}$  ages (less than a few tens of kyr) for a crystalline rock area. The obtained trend suggests that the deep groundwater in a sedimentary basin is relatively insusceptible to sea-level changes.

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## The study of the Torah scrolls from the National Museum of Brazil collection

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This study aims to support the critical analysis of the book of Deuteronomy, transcript in part of the nine scrolls of parchment deposited in the National Museum of Brazil collection. The text witnesses the five books of Torah, written in quadratic consonant Hebrew. Dom Pedro II, Emperor of the Second Brazilian Reign, possibly purchased the scrolls in his second trip to Europe from 1876 to 1877. Textual confrontation of the Deuteronomy writing fragments between ancient masoretic and late medieval copies was performed according to four stages of investigation : collation, analysis of the readings, study of textual family and paleography. Radiocarbon dating associated to Dead Sea Scrolls has been performed since the beginnings of Radiocarbon Dating and analysis of historical parchment was done for many samples since then (Brock F 2013 and references therein). Several chemical pre-treatments have been applied in order to remove contamination. Storage conditions were found to be very important. In this work we compared different treatments of the parchment and we dated each of the nine scrolls. Comparison between samples revealed not much variation among individual scrolls and no important contamination issues. The group of nine rolls results was considered as a Phase in the OxCal software and the historical boundary was used to limit the sequence. The results indicate the Scrolls are not older than the XVII century and the modelled time span ranges most probably from the XVIII to the XIX centuries. The results are in agreement with the textual analysis of the document.

References : Brock F. RADIOCARBON, Vol 55, Nr 2-3, 2013, p 353-363

Topic : GAA 93

Poster Session 2

## The $^{129}\text{I}$ -Iodine content of some seaweeds in Korea pre- and post-Fukushima nuclear accident

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The Tohoku earthquake and subsequent tsunami on 11 March 2011 resulted in serious damage to the Dai'ichi Fukushima Nuclear Power Plants ( $37^{\circ} 25' \text{ N}$ ,  $141^{\circ} 20' \text{ E}$ ), which released a broad suite of radionuclides into the environment via atmospheric plumes and direct discharge into the nearby ocean. Since the geographical distance between Japan and Korea is very close, it is highly necessary to monitor constantly the possible radioactive contamination in the Korean environment from the Fukushima accident by using various methods. In this study, we investigated the concentrations of  $^{129}\text{I}$  and the ratios of  $^{129}\text{I}/^{127}\text{I}$  in some seaweed samples collected from near Pusan ( $35^{\circ} 02' \text{ N}$ ,  $129^{\circ} 18' \text{ E}$ ), Korea by using accelerator mass spectroscopy (AMS). Samples were collected before and after the Fukushima nuclear accident. To our knowledge, there are scarce AMS data of the concentrations of  $^{129}\text{I}$  in seaweed samples from Korea that can be used by researchers to investigate the influence of Fukushima nuclear accident. Considering the high analyzing sensitivity of AMS, the collected data are expected to be used as good reference values for analyzing the impact of Fukushima disaster and the global cycle of the  $^{129}\text{I}$ .

Topic : GAA 94

Poster Session 2

### Determination of cross sections of $^{60}\text{Ni}(n,2n)^{59}\text{Ni}$ induced by 14MeV neutrons with accelerator mass spectrometry

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The production of long-lived  $^{59}\text{Ni}$  as activation product through  $(n,2n)$  reaction of 14MeV neutron on stable  $^{60}\text{Ni}$  is of concern for a fusion environment since they may lead to significant long-term waste disposal. However, their results of  $^{59}\text{Ni}$  via the  $(n,2n)$  reaction is strongly discordant, their results disagree by a factor of four. Such a discrepancy is far from the required accuracy needed for activation calculations in fusion reactor design technology. Based on the high sensitivity of  $^{59}\text{Ni}$  measurement at China Institute of atomic energy, determination of the cross section is being developed. Three natural nickel foils with a thickness of 0.2mm were irradiated on a D(T,a)n neutron generator, the energy of incident D beam was 300 keV, To avoid interference from thermal neutrons, the samples were wrapped with Cd foils during irradiation.  $^{57}\text{Co}$  and  $^{58}\text{Co}$  which produced by the  $^{58}\text{Ni}(n,np+pn+d)^{57}\text{Co}$  and  $^{58}\text{Ni}(n,p)^{58}\text{Co}$  reaction were chosen for the neutron flux determination. The neutron flux of  $(5.60 \pm 0.28) \times 10^{13}$  was determined by measuring  $\gamma$  ray emitted from  $^{57}\text{Co}$  and  $^{58}\text{Co}$  in the induced activity samples. After the neutron flux determination and Ni foil were dissolved, and NiO were made for AMS measurement. The amount of produced  $^{59}\text{Ni}$  will be measured via accelerator mass spectrometry utilizing the 13-MV tandem accelerator combine an Q3D magnet spectrometry of in China institute of atomic energy.

Work supported by the National Science Foundation of China, under Grant No. 11175266.

**Climate history of the Swiss Jura mountains derived from  $^{36}\text{Cl}$  in a limestone core****Alfimov Vasily,<sup>1</sup> Ivy-Ochs Susan,<sup>1</sup> Kubik Peter,<sup>1</sup> Beer Juerg,<sup>2</sup> Suter Martin,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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We have measured  $^{36}\text{Cl}$  concentration in a 100-m long limestone core from Vue des Alpes, Jura Mountains, Switzerland. The measurements were compared with our theoretical calculation of  $^{36}\text{Cl}$  content in the core. The long-lived radionuclide  $^{36}\text{Cl}$  ( $T_{1/2} = 301$  kyr) is produced in limestone by cosmic rays. There are several pathways of  $^{36}\text{Cl}$  production in the limestone. At the surface the dominant production pathway is spallation of Ca by fast neutrons. Below one meter of rock, the slow muon capture on  $^{40}\text{Ca}$  starts to dominate, while after 10 m depth the fast-muon-induced processes in Ca play a significant role. Additionally, three mentioned processes plus U-Th content of the rock produce thermal neutrons, and these neutrons activate stable  $^{35}\text{Cl}$  into  $^{36}\text{Cl}$ . These are also important pathways, because concentration of stable chlorine in the sampled core was non-negligible (65 ppm on average, 17-210 ppm the whole range). All production pathways were combined in a model of  $^{36}\text{Cl}$  production and applied to the calculation of  $^{36}\text{Cl}$  content of the core. With exception of two clear outliers, the model explained most of the depth profile. The main conclusion of this study is that the Last Glacial Maximum had negligible influence on Vue des Alpes, and the last major re-shaping of the landscape happened at Vue des Alpes about 140 kyr ago, when the glacier removed more than 15 m of rock.

Topic : GAA 96

Poster Session 2

## Radiocarbon determination of carbonaceous particles (organic carbon and elemental carbon) in rainwater samples

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Carbonaceous particles (CP), which comprise the large fractions of elemental carbon (EC) (also called black carbon; BC) and organic carbon (OC), badly affect climate and human health. The concentration and sources of CP in precipitation are important parameters for understanding of the detailed processes of wet deposition which is known to be a key scavenging (removing) process of OC and EC. Radiocarbon ( $^{14}\text{C}$ ) measurements of both OC and EC allow an improvement in carbonaceous aerosol source apportionment, leading to a full and unambiguous distinction and quantification of the contributions from non-fossil and fossil sources. However, such a method has not been applied to the precipitation samples. Here we develop a thermal-optical method with a commercial OC/EC analyzer to isolate water insoluble OC (WISOC) and EC of the filtered precipitation samples. The temperature protocol is optimized to separate OC and EC without interfering fractions with the best possible recovery. For their  $^{14}\text{C}$  determinations,  $\text{CO}_2$  resulting from the sample analysis is transferred to the gas ion source of the accelerator mass spectrometer MICADAS. The concentrations of WISOC and EC as well as their corresponding fraction of modern (fM) in rain samples collected in Dübendorf, Switzerland in 2012 will be measured. And the distinction and qualification of the biogenic and anthropogenic sources of particles in precipitation as well their seasonality will be discussed.

Topic : GAA 97

Poster Session 2

**Study on calcium absorption rate of rats by  $^{41}\text{Ca}$  labeling endogenous calcium****Du Liang,<sup>1</sup> He Ming,<sup>1</sup> Mi Shengquan<sup>1,2</sup> Pang Fangfang,<sup>1</sup> Wang Xiaoming,<sup>1</sup> Yang Xuran,<sup>1</sup> Zhao Qingzhang,<sup>1</sup> Jiang Shan.<sup>1</sup>**

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Calcium is one of the important elements that form human bone (the main form of Calcium is  $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ ). It participates in and regulates many life processes. Osteoporosis is the most common disease of calcium deficiency. It is a serious threat to human health, especially for old people, but recent research shows that organisms (especially osteoporosis organisms) take in too much calcium, possibly causing some other diseases. Thus, the accuracy of calcium absorption rate measurement is very meaningful for reasonable calcium supplement and prevention diseases of calcium metabolism. Calcium isotope tracer technology is an effective method to study the biological effects of calcium. As the best tracer of all calcium isotopes,  $^{41}\text{Ca}$  can only be tested through accelerator mass spectrometry (AMS). Based on the high sensitivity of  $^{41}\text{Ca}$  measurement with accelerator mass spectrometry and the innovative methods of  $^{41}\text{Ca}$  labeling endogenous calcium, the calcium absorption rate of rats will be studied in this work.

This work was supported by the National Natural Science Foundation of China (No. 11375272)

Topic : GAA 98

Poster Session 2

## Paleoclimatic study of the Gouveia region, Minas Gerais, Brazil, through Carbon isotopes and phytolith analysis

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In this work we study the chronology of a gully, a landform created by running water eroding sharply into soil typically on a hillside, located in the Gouveia region, in the Southern Espinhaço Mountain Range, Minas Gerais State, Brazil. We aim to associate phytolith and carbon isotopes analysis in order to better understand the evolution of the climatic conditions that influenced the geomorphic processes operating in the region during the Pleistocene / Holocene. For this study 13 samples were collected, with depths ranging from 30 cm to 7.30 m. The SOM fraction of samples was dated by  $^{14}\text{C}$ -AMS at the Radiocarbon Laboratory of the Fluminense Federal University using a 250 kV Single Stage AMS system. The results cover the last 40 ky with some age inversions. Isotopic analysis show the dominance of C4 plants in all samples, and the samples from depths between 5.20 and 6.20 m are the most  $^{13}\text{C}$  depleted. These same samples are the most enriched in carbon and also those with the greatest amount of phytoliths. The presence of Poaceae phytoliths was observed, with a decrease in the amount of short cells types and an increase of the bulliform type with depth.

Topic : GAA 99

Poster Session 2

## Accelerator mass spectrometry analysis of $^{14}\text{C}$ -oxaliplatin concentrations in biological samples and antineoplastic agents

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Abstract Microdosing studies have been proposed as means of obtaining human pharmacokinetics information at early stages of drug development. Accelerator mass spectrometry (AMS) has high detection sensitivity and has been used in the fields of archaeology, environmental science, and geology. In this study, we measured the  $^{14}\text{C}$  concentration in  $^{14}\text{C}$ -oxaliplatin-spiked biological samples. The calibration curves of  $^{14}\text{C}$  concentration in serum, urine and feces were linear, and the correlation coefficients were  $\geq 0.9893$ . The mean background  $^{14}\text{C}$  concentration in urine samples of 6 healthy Japanese volunteers was 0.144 dpm/mL, and the coefficient of variation in urine was higher than that in blood or plasma. The intra-day fluctuation of  $^{14}\text{C}$  concentration in urine from a volunteer was 15.3%. The antineoplastic agents are administered to the patients in combination. Therefore, quantitating background  $^{14}\text{C}$  concentrations of the antineoplastic agents is important.  $^{14}\text{C}$  concentrations were different among 10 antineoplastic agents;  $^{14}\text{C}$  concentrations of paclitaxel injection, docetaxel hydrate injection and irinotecan HCl hydrate injection were higher than those of the other injections. These results indicate that our AMS-based quantitation method is suited for microdosing studies and that measurement of baseline and co-administered drugs is necessary for the study.



## AMS Dating of the Danube fluvial terraces in the Romanian Plain

Enachescu Mihaela,<sup>1</sup> Stan-Sion Catalin,<sup>1</sup> Constantin Florin,<sup>1</sup> Enciu Petru,<sup>2</sup> Simion Corina Anca,<sup>1</sup>  
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Landscape evolution is the result of the interaction between tectonics, trying to create topography and climate driven surface processes. The Danube is the only river cutting through the Hungarian and Romanian Mountain Range offering the opportunity to determine its uplift rate via incision rates derived from terrace chronology. Unfortunately, the classical determination methods existing in geology have lead to contradictions. Therefore, the application of AMS dating method for million of years, based on  $^{10}\text{Be}/^{26}\text{Al}$  ratios measurements [1,2], is expected to give precise and confident age values of traces formation. Samples were collected from selected terraces and depth values. AMS experiments are carried out by use of the new Cockcroft Walton type 1 MV HVEE tandetron AMS system [3] recently installed at the laboratory in Bucharest. The stable isotopes ( $^{27}\text{Al}$  and  $^9\text{Be}$ ) were measured by ICP-MS. The AMS results obtained do not exceed 1% relative standard deviation. The work will present experimental determined age values for terraces and model calculations for their formation and of incision rates along the river.

### References :

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[3] C. Stan-Sion, M. Enachescu, D.G. Ghita, C.I. Calinescu, A. Petre, D.V. Mosu, M. Klein, NIM B 319 (2014) 117-122

Topic : GAA 101

Poster Session 2

**Using Radiocarbon in coral skeletons to reconstruct seawater pH at Milne Bay, PNG****Fallon Stewart,<sup>1</sup> Fabricius K.<sup>2</sup>**

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Porites coral cores have been collected from unique volcanic CO<sub>2</sub> seeps in Milne Bay Province, Papua New Guinea. The CO<sub>2</sub> gas bubbles emerging from the reefs provide local ocean acidification conditions similar to those predicted for the middle to the end of this century, and beyond. Volcanic CO<sub>2</sub> bubbling through the seawater in Milne Bay is free of radiocarbon, resulting a unique signal that is preserved in the coral skeleton. We have measured the radiocarbon content of the coral skeleton back through time from sites heavily impacted by CO<sub>2</sub> and "control" sites not impacted by CO<sub>2</sub> seeps. Three impacted sites show an increase of CO<sub>2</sub> into the DIC by 4%, 10% and 14%. Using these values we can estimate the pH at the impacted sites. In 2009 the impacted sites had estimated pH of 7.85, 7.6 and 7.4. These values agree with in situ measurements of seawater pH at the time the corals were collected.

## Glaciation history of Queen Maud Land (Antarctica) - new exposure data from Nunataks

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Rock exposure ages to cosmic radiation for the Wohlthat Massiv (Antarctica), had previously been analysed. This was done using quartz rich samples for  $^{10}\text{Be}$  and  $^{26}\text{Al}$  measurements by accelerator mass spectrometry at the AMS facility in Zürich. In order to determine the extent to which the results from the Wohlthat Massiv are of regional significance, additional samples were collected during the 2007 BGR-expedition "Queenmet". Two of the Steingarden Nunataks (isolated mountain peaks) were chosen as sampling locations, approximately 100 km south-east of the Wohlthat Massiv/Queen Maud Land, at the edge of the polar plateau. Quartz rich samples were collected at different elevations of the Nunataks to reconstruct an elevation-dependent exposure history. The in-situ produced cosmogenic nuclides  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in these samples were measured by AMS. The quartz separates were prepared by two different methods (Kohl und Nishiizumi 1992, Altmaier 2001) and measurements were performed at two different facilities (CologneAMS und Zürich AMS) to confirm the reproducibility of the results. The new results on exposure of rock surfaces reveal that the exposure of the lower Nunatak to cosmic radiation started 0.65 to 1.1 My ago, while the higher regions of the second Nunatak were apparently above the ice 3 to 4 My ago. A comparison of the different preparation procedures as well as a detailed discussion of the exposure data with respect to glaciation history of Antarctica will be presented.

Topic : GAA 103

Poster Session 2

**New software for AMS data analysis developed at IF-UFF Brazil****Diaz Castro Maikel<sup>1,2</sup> Chaves Damasio Macario Kita,<sup>1</sup> Silveira Gomes Paulo Roberto,<sup>1</sup>**

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A new software for AMS data analysis, named LACAMS, has been developed for determination of radiocarbon ages from AMS accelerator data. Written in C++ and using Qt libraries, it was developed to be used in the most common operating systems : Windows, Linux and OS X. This program, with a friendly graphical user interface, allows run discrimination, cathode grouping, standard sample and background source selection. In addition, several options can be configured to make more flexible the  $\delta^{13}\text{C}$  corrections and sample normalization, including run-by-run corrections, sample corrections and pre-normalization options. For every analysis, the whole dataset, samples, analysis options and results can be saved like a project, what makes it easy to continue or to modify the analysis at anytime. Results can also be exported using HTML format, which can be open with any browser. LACAMS also allows to make analysis for other AMS isotopes besides  $^{14}\text{C}$  and, for a major portability, it uses a plugins system allowing to load almost any dataset and therefore to analyze data from a wide range of AMS facilities.

Topic : GAA 104

Poster Session 2

**Dynamics of marine sediments studied through  $^{10}\text{Be}$** 

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Marine sediments may originate from the erosion of continental material (containing both cosmogenic  $^{10}\text{Be}$ , and  $^9\text{Be}$  with a ratio around  $10^{-8}$ ) that has been carried by rivers to the sea. If the sediments are deposited in zones where a tectonic plate subducts beneath another one, they might follow complex processes, in which part of the sediments are dragged under the plate and the other part is accreted above. In this work, depth profiles of the  $^{10}\text{Be}/^9\text{Be}$  ratios in marine sediments are being studied near the spot where Nazca, Antarctica and South American tectonic plates join each other. A preliminary set of seven samples, provided by the Ocean Drill Project [1], were measured at the DREAMS facility [2]; this represents the first measurement of a depth profile near this zone. The isotopic ratios, based on AMS-measurements of  $^{10}\text{Be}/^9\text{Be}$  and determinations of  $^9\text{Be}$  concentration performed by ICP-MS at HZDR are ranging from 4.9 to  $53 \times 10^{-9}$ . Contrary to the expectation they do not decrease with depth, but rise into the interval corresponding to 102 to 145 meters of depth, and from 197 to 256 meters of depth. We show that this result is consistent with a reverse (thrust) fault in the sediments due to the compression pressure exerted by the subduction of the Nazca tectonic plate.

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## New radiocarbon dates on upper mid-west proboscideans : determining date robustness.

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With the objective of refining the picture of Megafaunal extinction patterns in the upper Midwest in the terminal Pleistocene, we have assembled for radiocarbon dating specimens from more than 80 distinct Mammut and Mammuthus remains from potentially late sites. Measurements for this project will nearly double the extant number of published dates. These new specimens were all from museums rather than excavation sites, and 60% were known to be coated with a consolidant. The predominant consolidant was Butvar B-76, however shellac, Elmer's Glue, Glyptol were also noted in the conservation records, or deduced from knowledge of a particular museum's practices. Given the objective of the project is to identify extinction patterns, coupled with the wide prevalence of consolidants amongst the specimen set, it was imperative that extensive testing was carried out so that the dates can be considered robust. To this end, key specimens were dated three times using different sample preparation protocols. These were 1) a solvent extraction followed by a modified Longin-plus -Base continuous flow collagen extraction method used in the NSF-Arizona AMS facility, 2) the solvent/modified Longin method plus ultrafiltration, and 3) solvent/modified Longin method plus hydroxyproline single amino acid dating. Among the specimens subjected to triplicate testing were some of the youngest late Wisconsin proboscidean specimens from the Upper Midwest Region. The data reveal general agreement between the different protocols, and suggested either limited penetration of consolidants into the specimens, or that the standard laboratory cleaning protocols were sufficient to remove traces from deep within bone, tooth or tusk tissue. The preservation of each specimen, recorded in terms of collagen content, C/N ratio and stable isotope values, indicated that most were actually well preserved, implying the application of consolidant in the first place might have been unnecessary. The implications of these measurements, in terms of elucidating megafaunal extinction patterns, will be presented in future publications.

## Source apportionment of atmospheric PAHs from Kolkata, India by using compound class specific radiocarbon analysis (CCSRA).

Hidetoshi Kumata,<sup>1</sup> Masao Uchida,<sup>2</sup> Mahua Saha<sup>3</sup> Rina Kurumisawa,<sup>3</sup> Shoichi Saito,<sup>1</sup> Tomonari Umemura,<sup>1</sup> Miyuki Kondo,<sup>2</sup> Yasuyuki Shibata,<sup>2</sup> Tomoaki Okuda,<sup>4</sup> Fumiyuki Nakajima<sup>5</sup> Hideshige Takada.<sup>3</sup>

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Polycyclic aromatic hydrocarbons (PAHs) in the air originate mostly from combustion of organic materials. PAHs account for most of the total mutagenic activity of atmospheric aerosols. Hence, reducing air pollution by PAHs is essential for public health, which requires reliable source apportionment. Atmospheric pollution by PAHs in Indian megacities is comparable to the highest levels across the globe and Kolkata air exhibit the highest level among them [1]. This study aimed to apportion sources of combustion to atmospheric PAHs in Kolkata city and surrounding rural sites by using both source diagnostic PAH ratios and compound class specific radiocarbon analyses (CCSRA). Preliminary analysis of TSP aerosols revealed the significantly higher PAHs concentrations in urban sites (15-266 ng/m<sup>3</sup>) compared to the rural sites (2.5-61 ng/m<sup>3</sup>). Molecular fingerprinting gave basically the same source information for both sites. That is, combustion of coal in brickyards, wood for cooking, and diesel-soot to be major combustion to TSP-bound PAHs. To achieve more detailed source diagnosis, three- and four ring PAHs (MW178, 192, 202) in TSP samples from those two sites were isolated by using preparative-capillary-GC and analyzed for radiocarbon (<sup>14</sup>C) on AMS at NIES-TERRA, NIES (Tsukuba, Japan). The <sup>14</sup>C-based source apportioning between fossil and contemporary carbon fuels will be discussed in the presentation.

[1] doi : 10.1016/j.atmosenv.2013.03.001

### Upgrading of Beijing HI-13 tandem accelerator injector system.

Li Kangning,<sup>1</sup> You Qubo,<sup>1</sup> Bao Yiwen,<sup>1</sup> Guan Xialing,<sup>1</sup> Hu Yueming,<sup>1</sup> Su Shengyong,<sup>1</sup> Huang Qinghua,<sup>1</sup> Wang Xiaofei,<sup>1</sup> Kan Chaoxin,<sup>1</sup> Yang Tao,<sup>1</sup> Fan Hongsheng,<sup>1</sup> Yang Baojun,<sup>1</sup> Liu Dezhong,<sup>1</sup> Yang Bingfan,<sup>1</sup> Jiang Shan,<sup>1</sup> He Ming,<sup>1</sup> Kejun Dong,<sup>1</sup> Weiping Liu,<sup>1</sup> Renwei Hu,<sup>1</sup> Yin Ren,<sup>1</sup> Zhengyu Ma,<sup>1</sup> Xiuhua Zhang,<sup>1</sup> Fang Yan,<sup>1</sup> Qiuju Wang,<sup>1</sup> Minglong li.<sup>1</sup>

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Thirty years have past since the Beijing HI-13 tandem accelerator became operational at China Institute of Atomic Energy (CIAE) in 1984. The original injector consists of a trim lens and a 90°double-focusing analyzing magnet with a mass resolution ( $M/\Delta M$ ) of about 80, far from the required for Accelerator Mass Spectrometry (AMS) measurement of heavy nuclides and upgrading of HI-13 tandem accelerator. In recent years, the accelerator injector system was upgraded and optimized step by step in order to meet user's requirements. As a result, a dedicated AMS injection beam line with high mass resolution and a superconducting energizer with double drift buncher were reconstructed. In this contribution, some renovations of Beijing HI-13 accelerator injector system and corresponding performance improvements will be briefly introduced.



Topic : ISSI 07

Poster Session 2

**Sputter-pits casting****Shanks Richard,<sup>1</sup> Freeman Stewart.<sup>1</sup>**

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Sample-use efficiency is an important AMS parameter. Improvements promote increased counting statistics and the potential to reduce sample size or carrier added. Casting of the pit in Cs<sup>-</sup>sputtered targets has been done to measure primary-beam focus and to asses the effects of varying this, through ion source geometry modifications, on sample longevity, secondary-beam current and overall efficiency. The technique demonstrated here can aid in the optimisation of an ion source for maximum performance AMS.

Topic : ISSI 08

Poster Session 2

**Simultaneous and precise  $^{13}\text{C}$  and  $^{14}\text{C}$  measurements of gas samples****McIntyre Cameron,<sup>1</sup> Wacker Lucas,<sup>1</sup> Fahrni Simon,<sup>1</sup> Eglinton Timothy.<sup>1</sup>**

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Samples analyzed for radiocarbon in global carbon cycle studies require high precision  $^{13}\text{C}$  measurements to help interpret individual processes.  $^{13}\text{C}$  measurements on the MICADAS system at ETH Zürich have a precision of 2-3 permil which is sufficient for the correction of  $^{14}\text{C}$  ratios but too low for our geochemical and biogeochemical samples. A new stable isotope mass spectrometer (IRMS) has been purchased for integration with an elemental analyzer and the gas ion source of the MICADAS system. This will enable high precision  $^{13}\text{C}$  and  $^{14}\text{C}$  measurement to be made on bulk sedimentary samples and individual compounds. Will we present the modes of integration of the stable (IRMS), performance results and future prospects of the system.

Topic : ISSI 09

Poster Session 2

**Direct injection of carbon dioxide from headspace vials into a gas ion source****Seiler Martin,<sup>1</sup> Fahrni Simon,<sup>1</sup> Gautschi Philip,<sup>1</sup> McIntyre Cameron,<sup>1</sup> Wacker Lukas,<sup>1</sup> Synal Hans-Arno.<sup>1</sup>**

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Radiocarbon measurements on gas samples are routine at ETH Zürich with more than 1500 samples measured in 2013. With our current gas handling system, a zeolite trap is used to trap and transfer CO<sub>2</sub> to a syringe from a sample combustion or carbonate decomposition system. A constant flow of CO<sub>2</sub> gas flow is then introduced into the source as a 5% mixture in Helium. While this procedure is efficient and produces blanks better than 45K years, it requires a long routine of steps and has a cross contamination between samples of less than 1%. Measurements could be facilitated and cross contamination reduced if the gas flow to the ion source would come directly from a sample container. As carbonate samples can be easily converted to carbon dioxide by decomposing them with phosphoric acid in He flushed septa sealed vials, we have implemented a method that directly flushes the gas mixture from a septa sealed vial. We will present the technical realization of the direct injection of gas samples containing as little as 100 µg carbon or even less. A comparison of advantages and disadvantages of the direct injection of CO<sub>2</sub> without zeolite trapping and syringe injection will be presented.

Topic : MNSI 02

Poster Session 2

## Towards improvement in Al assay in quartz for in situ cosmogenic $^{26}\text{Al}$ exposure dating and $^{26}\text{Al}$ - $^{10}\text{Be}$ burial dating

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[1]Institute for Environmental Research, ANSTO (Australia)

Precise, accurate measurement of Al concentrations in quartz ([Al]qz), extracted from surface bedrock/sediment, is critical to obtain reliable  $^{26}\text{Al}$  exposure ages and  $^{26}\text{Al}$ - $^{10}\text{Be}$  burial ages. The [Al]qz is analysed by small aliquots, extracted from quartz-digested solutions, via, e.g. ICP-OES. Al loss during aliquot preparation, or inaccurate/inefficient assay during ICP-OES analysis can lead to erroneous [Al]qz assay and thus inaccurate  $^{26}\text{Al}$  ages or fictitious burial ages. At ANSTO, Al aliquots are processed in-house and Al analyses are carried out at labs both external and within ANSTO using ICP-OES. A 5-year analysis of [Al]qz variability in a "glass sand" powder (NIST SRM 165a; recommended [Al]  $312 \pm 13$  ug/g, 1s) shows a  $\sim 3\%$  variability with a long-term average  $283 \pm 8$  ug/g (1s,  $n = 25$ ),  $\sim 10\%$  lower than the certified value. A similar long-term study using an in-house purified quartz powder from a geological sample (OZ-2402) also shows a comparable, but somewhat elevated, long-term variability  $\sim 4.5\%$ . The observed variability 3-5% is higher than the 1% repeatability of duplicate Al aliquot solutions in the same batch. To investigate the cause of the large variability in [Al]qz assay and an apparent 10% offset in the [Al]qz value for the NIST-165a, we carried out tests on our existing aliquot preparation procedure, as well as standard addition method to test matrix effects in ICP-OES analysis. In this paper, we present results of these tests and discuss the reliability of [Al]qz assay via ICP-OES.

Topic : MNSI 03

Poster Session 2

## Data analysis at Leibniz Laboratory Kiel ; From AMS measurement to radiocarbon value.

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[1]Leibniz-Labor für Altersbestimmung und Isotopenforschung (Germany)

[2]Institute of Physics - Center for Science and Education (Poland)

[3]Centre for Baltic and Scandinavian Archaeology, Schleswig-Holstein State Museums Foundation, Schloss Gottorf, (Germany)

We have developed a method of correction for isotopic fractionation attributable to the ion source and the instability of the ion current for each sample. This is achieved by comparing the results for an unknown sample with results for NBS Ox II standard material with identical average values of the ion current for  $^{12}\text{C}$  and  $^{13}\text{C}$ . These values are obtained through fit-data function (ion current vs. isotopes ratio  $^{14}\text{C}/^{12}\text{C}$  and  $^{13}\text{C}/^{12}\text{C}$ ). Using this method it is possible to maintain high precision, even if the performance of the ion source is not stable during measurement. By applying this method we were able to decrease the scattering of the measurements.

Topic : NFF 12

Poster Session 2

**Status of the new AMS facility at the institute of applied physics, national academy of sciences of Ukraine.**

Moskalenko V.,<sup>1</sup> Boychenko A.<sup>1</sup> Buhay A.<sup>1</sup> Chivanov V.<sup>1</sup> Danylchenko S.<sup>1</sup> Drozdenko A.<sup>1</sup> Storizhko V.<sup>1</sup>

[1]Institute of Applied Physics, National Academy of Sciences of Ukraine, (Ukraine)

The accelerator-based mass spectrometer AMS 1.0 MV Tandetron manufactured by HVEE B.V. (Netherlands) has recently been put into operation at the Institute of Applied Physics, National Academy of Sciences of Ukraine (IAP NASU). The AMS facility is equipped with a S0110 hybrid ion source which permits analyses to be performed of both solid (graphite) and gaseous (CO<sub>2</sub>) samples. The machine is intended for measurements of cosmogenic radionuclides <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca, <sup>129</sup>I and also of transuranium isotopes Pu and U in geological, environmental, biological and pharmaceutical samples as well as in archeological artefacts. To provide the optimum AMS operation the necessary auxiliary equipment has been designed and constructed at the IAP NASU, viz. a system for drying and regeneration of the insulating gas (SF<sub>6</sub>), cooling water loop, etc. The AMS performance data obtained in the tests are the following : for the background isotope ratio <sup>14</sup>C/<sup>12</sup>C = 1.29×10<sup>-12</sup> the average statistical error is 0.384 %, relative standard deviation is 0.38 %, with the background being 2.28×10<sup>-15</sup>. The performance data of the newly installed equipment are comparable with the data of other AMS facilities.

### A New and Compact System at the AMS Laboratory in Bucharest.

Stan-Sion Catalin,<sup>1</sup> Enachescu Mihaela,<sup>1</sup> Ghita Dan Gabriel,<sup>1</sup> Simion Corina,<sup>1</sup> Petre Alexandru Razvan,<sup>1</sup> Calinescu Catalin Ionut,<sup>1</sup> Gaza Oana,<sup>1</sup> Mosu Vasile Daniel.<sup>1</sup>

[1]Horia Hulubei National Institute of Physics and Nuclear Engineering - IFIN HH, (Romania)

AMS research started in our National Institute for Physics and Engineering (NIPNE) in Bucharest more than 15 years ago [1]. A first AMS facility was constructed based on our multipurpose 9MV tandem accelerator and was upgraded several times [2]. Applications using this home made machinery were performed using light nuclei like  $^2\text{H}$ ,  $^3\text{H}$  and  $^{26}\text{Al}$  and are still performed with important result for material science. Heavier isotopes were also used in research, but experiments were performed at the laboratory of our partners from the Technical University, Germany. In May 2012 a new Cockcroft Walton type 1 MV HVEE tandetron AMS system, was commissioned [3]. The results of the acceptance test of this new machine will be presented together with latest results. They will demonstrate the high efficiency of the AMS machine in terms of accuracy, precision and low background level, routine  $^{14}\text{C}$  age dating and of measurements of other radioisotopes ( $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{41}\text{Ca}$ ,  $^{129}\text{I}$  and  $\text{Pu}$ ). Two chemistry laboratories were constructed and are routinely performing the target preparation for carbon dating and for other isotope applications for geology, environment physics, medicine and forensic physics.

[1] Stan-Sion C, Ivascu M, Plostinaru D, Catana D, Marinescu L, Radulescu M, Nolte E, Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms 172, (2000), 29-33

[2] C. Stan-Sion, M. Enachescu, O. Constantinescu, M. Dogaru, Nucl. Instr. Meth. B 268 (7-8) (2010) 863-866

[3] C. Stan-Sion, M. Enachescu, D.G. Ghita, C.I. Calinescu, A. Petre, D.V. Mosu, M. Klein, Nuclear Instruments and Methods in Physics Research B 319 (2014) 117-122

Topic : NFF 17

Poster Session 2

**The First AMS Facility in Africa at iThemba LABS in Gauteng****Mullins Simon,<sup>1</sup>**

[1]iThemba LABS, (South Africa)

Accelerator-Based Sciences (ABS) in South Africa are based in two provinces, namely the Western Cape and Gauteng. The iThemba Laboratory for Accelerator Based Sciences (LABS) has facilities in both provinces, where the Gauteng site has an EN Tandem Accelerator inherited from the University of the Witwatersrand. The accelerator systems have been fully refurbished, including a pelletron charging system. The Low Energy Injection System was commissioned in late 2012/early 2013 and now the High Energy Analysis System is under installation. Once completed and commissioned - as will happen over the next few months - this will be the first AMS facility on the continent of Africa. Benchmarking against top-ranked AMS facilities will be undertaken and a full research programme will be implemented as requested by numerous users - both national and international - who supported the completion of the facility.



**AMS measurements of  $^{10}\text{Be}$  at the CENTA facility****Jeřkovský Miroslav,<sup>1</sup> Steier Peter,<sup>2</sup> Priller Alfred,<sup>2</sup> Breier Robert,<sup>1</sup> Povinec Pavel,<sup>1</sup> Golser Robin.<sup>2</sup>**

[1]Comenius University (Slovakia)

[2]University of Vienna, Faculty of Physics, (Austria)

$^{10}\text{Be}$  is naturally produced in very low concentrations by interaction of galactic cosmic rays with the atmosphere, surface rocks, or with extraterrestrial objects. Very sensitive methods are therefore necessary for its detection. AMS is mainly limited by the stable isobar  $^{10}\text{B}$ , while the requirements for mass separation are the least stringent of all standard AMS isotopes. As the AMS line at the CENTA laboratory does not yet include a fully capable analyzing system, possibility was tested to measure  $^{10}\text{Be}$  using only a small switching magnet as the ion analyzer. The method for suppression of  $^{10}\text{B}$  was developed at the VERA laboratory and is based on a silicon nitride foil stack used as a passive absorber. The MC-SNICS was used for the production of  $^{10}\text{BeO}^-$  ions, which were mass separated and injected into the 9SDH-2 Pelletron, which operated at 3 MV terminal voltage.  $^{10}\text{Be}^{2+}$  ions were selected, and  $^{10}\text{B}$  ions as well as of most background ions from heavier masses were absorbed in the silicon nitride stack. An ionization chamber with two cathodes, based on a design of the ETH Zürich, was used for the ion detection. Using this setup, a detection limit of the order of  $10^{-12}$  for  $^{10}\text{Be}/^9\text{Be}$  was achieved, which was mainly determined by scattering of  $^9\text{Be}^{2+}$  ions on residual gas inside the switching magnet.

Topic : NFF 19

Poster Session 2

## Retrospective study of $^{14}\text{C}$ concentrations in tree rings at the vicinity of the Jaslovské Bohunice NPP using the AMS technique

Jeřkovský Miroslav,<sup>1</sup> Povinec Pavel,<sup>1</sup> Steier Peter,<sup>2</sup> Šivo Alexander,<sup>1</sup> Richtáriková Marta,<sup>1</sup> Golser Robin.<sup>1</sup>

[1]Comenius University (Slovakia)

[2]University of Vienna, Faculty of Physics, (Austria)

The Department of Nuclear Physics and Biophysics of the Comenius University has a long tradition in radiocarbon measurements at different regions of Slovakia, focusing mainly on the impact studies of nuclear power plants (NPP) on the environment. The atmospheric radiocarbon has been monitored around the Jaslovské Bohunice NPP since 1967 by static absorption of  $\text{CO}_2$  in NaOH solution and gas proportional counting. In 2012, tree ring samples were collected using an increment borer at areas surrounding the Jaslovské Bohunice NPP. Each annual tree ring was identified, and graphite targets were produced for accelerator mass spectrometry (AMS) analysis, which were carried out at the VERA laboratory using the 3 MV Pelletron accelerator (NEC). The radiocarbon data obtained from the tree ring samples are in a reasonable agreement with annual  $^{14}\text{CO}_2$  atmospheric data averaged from monthly radiocarbon measurements.

Topic : NFF 20

Poster Session 2

## **RICH - A new AMS facility at the Royal Institute for Cultural Heritage, Brussels, Belgium.**

Boudin Mathieu,<sup>1</sup> Van Strydonck Mark,<sup>1</sup> Synal Hans-Arno,<sup>2</sup> Wacker Lukas.<sup>1</sup>

[1]Royal Institute for Cultural Heritage (Belgium)

[2]Laboratory of Ion Beam Physics (Switzerland)

Since 1989 the radiocarbon dating lab has their own graphitization system for  $^{14}\text{C}$  AMS dating but RICH did not possess their own AMS and measurements were carried out in collaboration with other AMS facilities. In April 2013 the Micadas AMS was installed at the Royal Institute for Cultural Heritage in Brussels and after 1 year operation the high stability of the Micadas can be proven. For individually measured samples we are able to reach an uncertainty level of less than 5 per mil while for repeated samples a precision better than 3 per mil was obtained. Unknown samples were also measured on the RICH-Micadas and on other AMS systems and the obtained results showed a good agreement.

Topic : NFF 21

Poster Session 2

## PIS-AMS-OQUAD - Portable in-situ accelerator mass spectrometry for one in a quadrillion detection♠.

Right Everette,<sup>1</sup> Doe John,<sup>2</sup> Yellow Duck,<sup>3</sup> Bond Jamie.<sup>4</sup>

[1] IKIA (I Know It All) institute, (Neverland)

[2] Green-red SLP-company, (Mars)

[3] The Duck pond Lab, (Mexico)

[4] 007 SMA Lab, London, (UK)

Following the recent studies of our colleagues leading the scientific field of accelerator mass spectrometry (AMS), we have developed a set-up that can be used for the detection of all long-lived radionuclides and even stable isotopes in-situ in the field. The set-up is a classical AMS one : ion source, accelerator, detector. However, as its total weight is less than 2.5 kg, it is portable and can be transported to the location of interest. In fact, neither sample taking nor chemistry is needed before AMS analyses. A single person can take the so-called PISAMSOQUAD into his/her backpack and do AMS-analyses in-situ. The advantages are manifold : for those of us who hates hiking in the mountains, there is no need to go home with your backpack filled with heavy stones from rock fall boulders or lava flows. No feed for hammering, crushing, sieving and the never-ending chemistry work (for those who hate this part). PISAMSOQUAD uses a commercial red laser pointer (SLP<sup>®</sup> = SuperLaserPointer, excitation) for extraction of the nuclides out of the surface layer and first ionisation. A 500 ml thermo flask (from the Alaska company FIB<sup>®</sup> = Freezing Ice Bear) filled with sugar-free ice-tea (specially brewed by Lipton) is used as an ion trap for preenrichment. After about 1min of collecting ions, the ions can be released to the accelerator originally build by Matchbox for kids entertaining. The new state-of-the-art acceleration TM system contains of an Y-shaped inverse double ski-jump for first isobar suppression. Total molecule destruction is guaranteed by guiding ions from a sophisticated small looping (diameter : 20 cm ; also from Matchbox ) for post-acceleration trough natural honeycombs that had been selected and collected near TM lavender fields by AMS-experts from ASTER, Aix-en-Provence. The detection unit is an human interface who transforms by hand each count (we call it character) from a CNRS-IT-solution-run system into a real pdf-document. The detection limits for all isotopes - including noble gases - are exceptional : One in a quadrillion. The whole system can be run by an iPod (Apple Inc.). A free beta version of the associated App, that has the only disadvantage of having no formatting options like superscript or subscript, which is usually not necessary for AMS-research - can be downloaded at [www.cerege.fr/PISAMSOQUAD](http://www.cerege.fr/PISAMSOQUAD) until the 1 of April st 2015. The whole system will be sold after the concluding remarks of AMS-13 to the potential organisers of AMS- 14 for  $1 \times 10^{12}$  euros. Finally, our first tests upgrading the system with a violet laser pointer (VSLP) for simultaneous speciation analysis at fault scarps of Sainte Victoire and Tsunami-triggered rock fall boulders from the cliff at Cassis look promising. We are proud to show the PISAMSOQUAD at AMS-13.

♠ : Abstract received the First of April from the SM German company.

## Evaluation of Intracavity Optogalvanic Spectroscopy at Uppsala University

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In 2008, the first report of an ultrasensitive method for ro-vibrational spectrometry of radiocarbon dioxide was published by Murnick et al. The method, called intracavity optogalvanic spectroscopy (ICOGS), claimed a sensitivity and limit-of-detection (LOD) comparable to AMS. ICOGS utilizes the narrow linewidth, isotope-dependent ro-vibrational absorption lines of carbon dioxide in the IR spectrum. Here, the lines of carbon dioxide molecules with different carbon isotopes are separated by several hundred linewidths. In order to facilitate unambiguous detection of radiocarbon, the sample is placed inside the laser cavity of a radiocarbon dioxide laser. This intracavity approach was claimed to increase the sensitivity of the detection by seven orders of magnitude compared with traditional optogalvanic methods. However, despite the methodical and thorough efforts of at least five research groups worldwide, these claims have not been possible to confirm. As the first group to properly repeat the original experiments, we have during the last year reported serious deficiencies in the reproducibility of the original results. We found that ICOGS in its original embodiment suffers from considerable problems with the stability and reproducibility of the optogalvanic signal, and that misinterpretations of these uncertainties likely are the explanation for the extraordinary sensitivity in the original reports. Moreover, the previously reported Voigt profile-like line shape of the radiocarbon dioxide absorption lines could not be reproduced for radiocarbon concentrations in the 0.1-10 Modern range. In this report, we further discuss our results, and present a new approach for improved signal detection.

**Biomedical Accelerator Mass Spectrometry at Uppsala University : Progress Report****Salehpour Mehran,<sup>1</sup> Possnert Göran,<sup>1</sup> Håkansson Karl.<sup>1</sup>**

[1]Department of Physics and Astronomy, Ion Physics, Uppsala University (Sweden)

Biomedical Accelerator Mass Spectrometry (AMS) research at our laboratory is focused on radiocarbon tracing. The Uppsala University 5 MV Pelletron tandem accelerator has so far been used for all the biomedical applications. By the start of the AMS-13 conference, a new 200 kV tandem AMS system, the Green-MICADAS AMS developed at ETH Zürich, will have been installed. The latest performance data of the system will be presented. With the new system installed, the natural level samples (<2 Modern) will be completely separated from the higher activity  $^{63}\text{Ni}$ -labelled samples, by using a separate AMS system as well as a dedicated sample preparation laboratory.

A variety of collaborative biomedical AMS projects is currently being pursued and will be presented. Examples are : i) Ultra-small sample AMS. Latest results are presented for samples down to a few  $\mu\text{g C}$ , ii) Bomb peak dating of human DNA. A long term project is presented where purified and cell-specific DNA from various part of the human body including the heart and the brain are analyzed with the aim of extracting regeneration rate of the various human cells, iii) Bomb peak biological dating of various human biopsies, including human post-mortem amyloidosis proteins, iv) A clinical, phase-0 microdosing measurements using a  $^{63}\text{Ni}$ -labelled macromolecular drug (ca. 30000 Daltons) candidate is presented, and v) Forensic dating of teeth is also addressed.

Furthermore, an update is given on the Uppsala laser-based, radiocarbon intra-cavity optogalvanic spectroscopy system.

Topic : PRE 12

Poster Session 2

## A New Method for Beam Emittance Measurements : Construction of an Allison Scanner for the Erlangen AMS Facility

Stuhl Alexander,<sup>1</sup> Kainz Maximilian,<sup>1</sup> Scharf Andreas,<sup>1</sup> Schindler Matthias,<sup>1</sup> Kretschmer Wolfgang.<sup>1</sup>

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The Erlangen AMS facility had been installed at the pre-existing EN Tandem laboratory in Erlangen since the 1980s, so it is not a pre-designed AMS system that had been acquired as a whole, but an organically grown facility. In the context of major revisions on our beam transmission monitoring to keep it up-to-date, we have also tested a quite new method for beam emittance measurements of ion sources, a so-called Allison scanner. This high-precision measurement device consists of an electrical sweep plate between two plane-parallel slits which allows the simultaneous measurement of divergence and position of the beam. Together with the new beam transmission monitoring system we will present our first results of emittance measurements with a self-built Allison scanner and computer simulations for an upgraded MC-SNICS ion source. The simulations consist of a comparison between different simulation software and different mathematical procedures, such as the Finite Element Method (FEM), the Finite Difference Method (FDM), and the Boundary Element Method (BEM).

**Status report of NIES-TERRA : progress of 18 years' operation**

Uchida Masao,<sup>1</sup> Kobayashi Toshinobu,<sup>1</sup> Kondo Miyuki,<sup>1</sup> Mitsuguchi Takhiro,<sup>1</sup> Kato Fumiaki,<sup>1</sup> Shibata Yasuyuki.<sup>1</sup>

[1]National Institute for Environmental Studies (Japan)

The AMS facility (NIES-TERRA) at the National Institute for Environmental Studies has now been operating for seventeen years since 1996. The facility consists of a 5MV tandem Pelletron accelerator (NEC,15SDH-2) and a solid ion source with sequential injection system. Our main target is radiocarbon analysis in environmental sciences studies such as paleoclimate, carbon cycles, and atmospheric sciences, especially with development of various chemical applications such as compound specific  $^{14}\text{C}$  analysis. Most recently we started measurement of  $^{129}\text{I}$  as part of environmental studies associated with the Fukushima nuclear power plant accident. We saw successful throughput of measurements about 1000 samples per year including standards except for temporal shutdown since the 2011 mega-earthquake. To date our operational status is fortunately getting back to the condition before the shutdown. During the interval of the shutdown, the AMS system was refurbished by exchanges of a overall computer system for control and data acquisition for  $^{129}\text{I}$  analysis and various hardware including a new power supply of an analyzing magnet. However, recently we confirmed that the beam lines of the AMS (15SDH-2) was out of alignment by 3 mm between high and low energy system, which is most likely caused by the earthquake. This was seriously critical for  $^{129}\text{I}$  analysis, although beam adjustment for  $^{14}\text{C}$  analysis could be done with high precision as in the previous analyses before the earthquake. We have improved our preparation laboratory by semi-automatic graphitization lines to increase sample throughput. In the conference, we also present the perspectives of our facility for next decade for the various fields of environmental science studies.



Topic : PRE 14

Poster Session 2

**Ion-optics of the iThemba LABS AMS system****Sekonya Kamela<sup>1,2</sup> Sideras-Haddad Elias,<sup>2</sup> Brown Tom.<sup>3</sup>**

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[2]University of the Witwatersrand (South Africa)

[3]Lawrence Livermore National Laboratory (United States)

The 6 MV tandem accelerator of the iThemba LABS facility, which were mainly used for ion beam analysis has been modified for accelerator mass spectrometry. The beam optics of the 6 MV tandem accelerator at iThemba LABS is presented. Typical beam trajectories for proton and  $^{12}\text{C}$  beam are shown. The ion-optics calculation of low-energy injection beam line and high-energy beam line have been conducted.

## The first three years of DREAMS : Routine operation and developments

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The DREAMS (DREsden AMS) facility is based on a state-of-the-art 6 MV accelerator AMS-system [1]. Located at the ion beam centre, the accelerator is also used for ion beam analysis (IBA), material modifications and high-energy ion implantation. Though having no 24/7 availability for AMS, the advantage of a multi-purpose accelerator is the synergy effect with respect to joint technology development and  $\mu$ -beam IBA for the in-situ identification of elements in problematic cathodes. Most often measured nuclides are  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{36}\text{Cl}$ . The majority of samples is prepared on-site [2],  $^{36}\text{Cl}$  in a dedicated laboratory for halide targets. About 600  $^{10}\text{Be}$  unknowns have been measured for different applications [3]. The mean ratio of processing blanks is as low as  $3 \times 10^{-15} \text{ }^{10}\text{Be}/^9\text{Be}$ , even when measuring samples with ratios as high as  $10^{-10}$ - $10^{-11}$ . However, the mean machine blank is generally a factor of four lower. While analysing 150  $^{26}\text{Al}$  unknowns, machine blanks are as low as  $3 \times 10^{-15} \text{ }^{26}\text{Al}/^{27}\text{Al}$ . Typical high-energy currents ( $\text{Al}^{3+}$ ) e.g. for the in-house standard are about 300 nA (mean / 1 h). One of the original ion sources has been modified for reducing long-term memory for volatiles [4] and yet applied to  $\sim 100$  unknowns of  $^{36}\text{Cl}$  [e.g. 5]. Measurements of  $^{41}\text{Ca}$  are mainly for nuclear decommissioning [6] and cosmochemistry [5] applications. The high-energy setup is upgraded with a time-of-flight and energy detector system to perform actinide AMS and Super-SIMS [7].

[1] Akhmadaliev et al. NIMB 294 (2013) 5.

[2] Merchel et al. AMS-13.

[3] Feige et al., Ludwig et al., Ott et al., Rodrigues et al. & Smith et al. AMS-13.

[4] Pavetich et al. NIMB, (2014) 10.1016/j.nimb.2014.02.130 & AMS-13.

[5] Ott et al. MAPS, subm.

[6] Hampe et al. JRNC 296 (2013) 617.

[7] Rugel et al. AMS-13.

**MALT AMS system : current status and future direction**

**Matsuzaki Hiroyuki,<sup>1</sup> Nakano Chuichiro,<sup>1</sup> Tsuchiya Yoko,<sup>1</sup> Ito Seiji,<sup>1</sup> Morita Akira,<sup>1</sup> Kusuno Haruka,<sup>1</sup>  
Miyake Yasuto,<sup>1</sup> Honda Maki,<sup>1</sup> Bautista Angel Vii,<sup>1</sup> Kawamoto Marina,<sup>1</sup> Tokuyama Hironori.<sup>1</sup>**

[1]Micro Analysis Laboratory, Tandem accelerator, The University of Tokyo (Japan)

MALT (Micro Analysis Laboratory, Tandem accelerator, The University of Tokyo) is designed for highly sensitive and precise elemental and isotopic microanalysis system using ion beam generated by PelletronTM 5UD tandem accelerator. Currently multi-nuclide AMS ( $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{129}\text{I}$ ) system is available and shows good performance as well as PIXE, NRA, ERDA/RBS systems. The total operation time of the accelerator has been over 95,000 hours since the start of MALT, 20 years ago. After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, many projects related to  $^{129}\text{I}$  have been conducted. The retrospective reconstruction of  $^{131}\text{I}$  distribution at the accident from  $^{129}\text{I}$  is one of most important mission. So far more than 1,000 soil samples were analyzed and made a  $^{131}\text{I}$  distribution map. The accident derived  $^{129}\text{I}$  is also very useful as a tracer for the iodine dynamics in the environment. The contrast between  $^{127}\text{I}$  and  $^{129}\text{I}$  distribution not only in bulk soil but also in each soil fraction extracted by specific treatments tell us the useful information about the elemental processes taken in place. At MALT,  $^{129}\text{I}$  is detected by a gas ionization chamber and is highly sensitive. The background level is regularly monitored by measuring the "Old Iodine" provided from Woodward corporation and shows always  $^{129}\text{I}/^{127}\text{I} < 2 \times 10^{-14}$  as an AMS result. We are also trying to detect the accident derived  $^{36}\text{Cl}$  from soil samples. Current system at MALT uses a gas-filled magnet for the final detector. Recently we tried 6+ charge state instead of 7+ and confirmed adequate efficiency without degrade of sensitivity. For the environmental assessment related to the nuclear activity and accident,  $^{236}\text{U}$ -AMS system is now under development.

## Status of recent activities and operations at the national ocean sciences AMS (NOSAMS) facility.

Gagnon Alan,<sup>1</sup> Longworth Brett,<sup>1</sup> Roberts Mark,<sup>1</sup> Von Reden Karl,<sup>1</sup> McNichol Ann,<sup>1</sup> Gospodinova Kalina,<sup>1</sup> Jenkins William,<sup>1</sup>

[1]National Ocean Sciences Accelerator Mass Spectrometry Facility (United States)

For 23 years NOSAMS has supplied high throughput, high quality AMS  $^{14}\text{C}$  analyses to the ocean sciences community. We continue to operate two accelerators - a 3 MV Tandatron and a 0.5 MV compact AMS, both dedicated to radiocarbon. The Tandatron runs a wheel of 58 cathodes each operational day. We've replaced the recombinator with a sequential injector using a bounced magnet chamber to select isotopes for injection. This has reduced machine background by a factor of 2. We are working to replace the ion source with a 40 position NEC MCSNICS-II on loan from KCAMS at UC Irvine. This should better match the source output to the new injector and accelerator, increasing output and stability. The large-acceptance compact CFAMS (continuous flow AMS) has dual ion sources : a 134-position NEC MC-SNICS sputter and a microwave plasma gas ion source. The sputter source is used for all NOSAMS's small ( $< 300 \mu\text{g}$ ) samples, swipe and commercial samples, and for high-precision measurements. The gas ion-source is used for rapid, low-cost measurements on carbonate samples. We will install an increased diameter accelerator stripper canal, which should improve measurement precision using the gas source. In the sample preparation lab (SPL) samples to  $20\mu\text{gC}$  are routinely processed, and expanded analysis capabilities now include samples to  $5 \mu\text{g C}$ . We have developed a new method for extracting dissolved inorganic carbon from waters (REDICS). Isolating the carbon relies upon transfer across a polymer membrane. Pre-treatment of organic carbon samples is now accomplished with an automated system named the OCtoPuS. Our programmable ramped temperature pyrolysis/combustion system (the Ramped PyrOx) has been improved and being used frequently by faculty, students, and post-doc researchers.

Topic : PRE 18

Poster Session 2

**Two years since SSAMS : status of  $^{14}\text{C}$  AMS at CAIS****Prasad Gurazada,<sup>1</sup> Cherkinsky Alexander,<sup>1</sup> Culp Randy,<sup>1</sup> Dvoracek Doug.<sup>1</sup>**

[1]Center for Applied Isotope Studies (United States)

The NEC 250 kV single stage AMS accelerator (SSAMS) was installed two years ago at the Center for Applied Isotope Studies, University of Georgia. The accelerator is primarily used for radiocarbon measurements to test the authenticity of natural and bio-based samples whereas all other samples such as marine, geological, atmospheric and archaeological are run on the NEC 500 kV , 1.5SDH-1 model tandem accelerator, which has been in operation since 2001. Due to higher terminal potential, the tandem accelerator provides better precision, but the single stage unit requires much less maintenance. We discuss the operational parameters and compare the performance of both machines in this facility report.

Topic : PRE 19

Poster Session 2

### Progress on multi-nuclide AMS of JAEA-AMS-TONO

Saito-Kokubu Yoko,<sup>1</sup> Matsubara Akihiro,<sup>1</sup> Miyake Masayasu,<sup>2</sup> Nishizawa Akimitsu,<sup>2</sup> Ohwaki Yoshio,<sup>2</sup>  
Nishio Tomohiro,<sup>2</sup> Sanada Katsuki,<sup>2</sup> Hanaki Tatsumi.<sup>1</sup>

[1]Tono Geoscience Center, Japan Atomic Energy Agency (Japan)

[2]Pesco Corp. Ltd. (Japan)

The JAEA-AMS-TONO facility was established in 1997 at the Tono Geoscience Center, Japan Atomic Energy Agency (JAEA). Our AMS system is a versatile system based on a 5MV tandem Pelletron type accelerator (National Electrostatic Corporation, US) and has been made available for  $^{14}\text{C}$ - and  $^{10}\text{Be}$ -AMS. At present, the development of  $^{26}\text{Al}$ -AMS has been conducted to enhance the capability for multi-nuclide AMS. The AMS system has been mainly applied to dating in studies of neotectonics and hydrogeology, in support of our research on geosphere stability applicable to the long-term isolation of high-level radioactive waste. Furthermore, the  $^{14}\text{C}$ - and  $^{10}\text{Be}$ -AMS are used for geoscience, environmental science and archaeology by researchers of universities and other institutes under the JAEA's common-use facility program. The  $^{14}\text{C}$ -AMS is most dominant in utilization of the system and has used for radiocarbon dating since establishment of the facility. Routine  $^{10}\text{Be}$ -AMS started at the beginning of the fiscal year of 2013. In a next attempt to enhancement of the multi-nuclide AMS, we have started the development of  $^{26}\text{Al}$ -AMS. The  $^{10}\text{Be}$ - and the  $^{26}\text{Al}$ -AMS make us possible  $^{10}\text{Be}$  and  $^{26}\text{Al}$  dating which are effective to estimate the exposure age of basement rocks and the sedimentation rate. The system tuning and test measurement have been progressed and the routine measurement of the  $^{26}\text{Al}$ -AMS will be started in near future.

Topic : PRE 20

Poster Session 2

## Setup and first applications of a gas ion source for micro radiocarbon dating at the MICADAS-AMS system in Mannheim, Germany

Hoffmann Helene,<sup>1</sup> Kromer Bernd<sup>1,2</sup> Wagenbach Dietmar,<sup>1</sup> Fahrni Simon.<sup>3,4</sup>

[1]Institute for Environmental Physics (Germany)

[2]Klaus-Tschira-Laboratory for Archaeometry (Germany)

[3]ETH Laboratory of Ion Beam Physics (Switzerland)

[4]Ionplus AG (Switzerland)

For many dating problems in paleoclimate science (e.g. dating of Alpine glacier ice) the conventional graphite based AMS radiocarbon dating technique is not applicable. In these instances, available sample masses for radiocarbon dating are very small, typically in the microgram range of carbon mass and thus graphitisation is cumbersome. To make dating of such samples between 5 and 100 microgram carbon possible, an interface for direct CO<sub>2</sub> gas radiocarbon measurements (GIS), designed and built by the ETH Zurich / Ionplus AG, was installed and put into operation in early 2014 at the MICADAS (Klaus Tschira Laboratory for Archaeometry, KTA) in Mannheim. Here we report on the characteristics of the gas ion source system adapted to the settings of the AMS in Mannheim, including determination of optimal operational settings like caesium-temperature and CO<sub>2</sub>-mass flow to the ion source with regards to stable and high <sup>12</sup>C currents. Furthermore, investigations on radiocarbon contamination level and possible memory effects within the gas inlet system will be shown. Apart from the technical developments we will lay a strong focus on first scientific applications of the gas ion source system at KTA, mainly on the radiocarbon dating and age evaluation of the particulate organic carbon fraction obtained from selected Alpine glacier ice samples.

Topic : PRE 21

Poster Session 2

**Decadal  $^{10}\text{Be}$  and  $^{26}\text{Al}$  Measurements at the SUERC 5MV AMS Facility****Xu Sheng,<sup>1</sup> Freeman Stewart,<sup>1</sup> Rood Dylan,<sup>1</sup> Shanks Richard.<sup>1</sup>**

[1]Scottish Universities Environmental Research Centre, (United Kingdom)

The routine performance and uncertainties of decadal  $^{10}\text{Be}$  and  $^{26}\text{Al}$  measurements made on the SUERC 5 MV accelerator mass spectrometer are assessed. The analysis compiles data from primary (NIST reference SRM4325 for  $^{10}\text{Be}$  and Purdue Z92-0222 for  $^{26}\text{Al}$ ) and secondary (Nishiizumi's series for  $^{10}\text{Be}$  and  $^{26}\text{Al}$ ) reference samples with  $^{10}\text{Be}/^9\text{Be}$  and  $^{26}\text{Al}/^{27}\text{Al}$  ratios ranging between  $10^{-11}$  and  $10^{-13}$ . In general, our long-term datasets indicate that the six Be secondary standard samples have standard deviations from 1.1 % to 2.4 % with average difference from the nominal values from -0.4 % to 0 %, and that the Al series have standard deviations from 0.5 % to 2.5 % with average difference from the nominal values from 0.1 % to 1.0 %. Individually, the sample Be-01-6-2 with the lowest  $^{10}\text{Be}/^9\text{Be}$  ratio  $5.92 \times 10^{-13}$  has average statistical uncertainties based on counting statistics of 1.8 %. The mean measured ratio falls within the 0.4 % uncertainties of the nominal value. The standard deviation around the mean of this sample is 1.9 %. These data indicate an additional uncertainty (0.9 %) above that calculated from counting statistics alone. On the other hand, the sample Al-01-5-3 with the lowest  $^{26}\text{Al}/^{27}\text{Al}$  ratio  $4.99 \times 10^{-13}$  has average statistical uncertainties of 2.8 %. The mean measured ratio is within the 0.3 % uncertainties of the nominal value, and the standard deviation around the mean of this sample is 2.1 %. These show no additional uncertainty above that based on counting statistics alone.



Topic : PRE 22

Poster Session 2

**Status and laboratory report of the Seoul National University AMS****Jang Hoon Lee,<sup>1</sup> Chi Hwan Kim,<sup>1</sup> Jin Kang,<sup>1</sup> Sujin Song,<sup>1</sup> Myung-Ho Yun,<sup>1</sup> Jong Chan Kim,<sup>1</sup>**

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[2]School of Physics, College of Natural Science, Seoul National University (South Korea)

Seoul National University AMS : Tandetron 3 MV of HVEE has been operating nearly 10 years without a single tank opening despite that we had many openings in the period following the initial installation. We are now facing a problem of loosened timing belt at the motor side of the motor-generator (M-G) system. In this occasion of the tank opening after a long hiatus, some interesting observations such as state of diode chain stacks, resistance chains in the accelerator tubes, state of bearings in the M-G system, and the Ar stripper gas bottle level will be made and reported. In the middle of last year we had contamination problem in our combustion system utilizing an elemental analyser (EA). The contamination level (pMC > 155) cannot be explained by modern carbon intrusion and we suspect that highly  $^{14}\text{C}$  enriched carbon is mixed and contained in the high purity  $\text{O}_2$  which is used for EA system as oxidant, even though  $\text{O}_2$  and  $\text{N}_2$  gas bottles was replaced and connected to EA simultaneously. Oxygen is more likely candidate causing this problem since  $\text{N}_2$  was just used as a tool for the actuation of automatic sample insertion instead of a compressed air into the inlet of EA and we are investigating what stage of industrial process causes this problem.

## New Ion Source and Graphitization Line of YU-AMS

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[3]College of Art and Design, Musashino Art University (Japan)

[4]The Center of Academic Resources and Archives Botanical Gardens, Tohoku University, (japan)

[5] National Institutes for Cultural Heritage, Nara National Research Institute for Cultural Properties , (Japan)

In 2009, Yamagata University (YU) installed an AMS (YU-AMS) system in Kaminoyama Research Institute to meet the requirements of  $^{14}\text{C}$  AMS for microdosing and medical studies as well as those of radiocarbon dating in the same facility. The AMS system is based on a 0.5 MV Pelletron accelerator developed by National Electrostatics Corporation. This AMS system is the first AMS system installed in a university in northern Japan (Tohoku-Hokkaido region). The facility also provides radiocarbon dating for samples from other universities, institutes and public organizations. In March 2014, a second automated graphitization line and a second ion source on the AMS system were installed. The automated graphitization line can be used to treat more than 2,400 samples per year. The ion source can be used for a subsequent measurement by setting samples in a chamber and evacuating the chamber while the other source is being used for AMS measurement. Hence, this system can shorten the total measurement time. We carried out performance tests on the YU-AMS system by measuring the C series standard samples (C1 - C9) and HOxII provided by IAEA and NIST, respectively. In this conference, we describe the status of the YU-AMS system.

Topic : PRE 24

Poster Session 2

## Operation of the "Small" Spectrometers at CAMS Over the Past 13 Years : Past and Future Prospects

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[2]Biology and Biotechnology Division, Lawrence Livermore National Laboratory (United States)

Purchased in 1999, the LLNL 1-MV spectrometer was one of the first of the new smaller AMS systems and was designed to serve our biomedical research program. The system was configured with a copy of LLNL's high-output Cs sputter ion source coupled to an NEC tandem accelerator and analysis beamline. Operation of this system began in April 2001 and as of summer 2014, over 80,000 samples have been measured with the vast majority of those samples in support of the NIH-funded National Resource on Biomedical Accelerator Mass Spectrometry Research Resource. In 2009, we expanded the capabilities of this system with the installation of a second ion source and injection beamline. The second source is a heavily modified NEC MCGSNICS and is designed to ionize both solid and gaseous targets. The injection beamline was configured to allow either the simultaneous measurement of  $^3\text{H}/^1\text{H}$  ratios from solid  $\text{TiH}_2$  targets or the measurement of  $^{14}\text{C}/^{12}\text{C}$  from solid graphite or  $\text{CO}_2$  samples. The gas-ionization capabilities of this new ion source were not fully exploited until the installation of a moving wire interface in 2011. This interface enables the analysis of small liquid samples either as discrete microliter-sized drops or directly coupled to the output of an HPLC. We are further expanding our Liquid Sample AMS capabilities by building two more copies of our moving wire interface for deployment on a soon-to-be installed 250 kV SSAMS.

Work performed at the Research Resource for Biomedical AMS, which is operated at LLNL under the auspices of the U.S. DOE under contract DE-AC52-07NA27344, is supported by the National Institutes of Health, National Institute of General Medical Sciences, Biomedical Technology Research Resources under grant number 8P41GM103483.

Topic : PRE 25

Poster Session 2

**AMS measurement of  $^{53}\text{Mn}$  and its application at CIAE**

**Dong Kejun,<sup>1</sup> He Ming,<sup>1</sup> Liu Guangshan,<sup>2</sup> Hu Hao,<sup>1</sup> Dou Liang,<sup>1</sup> Liu Jiancheng,<sup>1</sup> Wu Shaoyong,<sup>1</sup>  
Wang Xianggao,<sup>3</sup> Shen Hongtao,<sup>4</sup> Li Kangning,<sup>1</sup> You Qubo,<sup>1</sup> Bao Yiwen,<sup>1</sup> Hu Yueming,<sup>1</sup> Jin  
Chunsheng,<sup>1</sup> Yin Xinyi,<sup>1</sup> Jiang Shan,<sup>1</sup>**

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[3]College of Physics Science and Technology, Guangxi University, (China)

The determination of cosmogenic  $^{53}\text{Mn}$  in terrestrial formations has important applications. Accelerator Mass Spectrometry (AMS) is the most sensitive technique to detect minute amounts of  $^{53}\text{Mn}$ .  $^{53}\text{Mn}$  measurement techniques have been developed in the past few years on the  $\Delta E$ -Q3D-equipped AMS system at China Institute of Atomic Energy (CIAE). The method has recently been further optimized with the goal for AMS measurement of  $^{53}\text{Mn}$  concentrations in a deep sea ferromanganese crust (DSFC) sample. Based on these improvements,  $^{53}\text{Mn}$ -containing in different depth profiles of DSFC were analyzed by AMS. The newest experimental progress, performances and results are detailed in this presentation.

### Progress in AMS Measurement of U Isotope Ratios in Nanogram U Samples

Dong Kejun,<sup>1</sup> He Ming,<sup>1</sup> Wang Chen,<sup>1</sup> Zhao Xinhong,<sup>1</sup> Li Lili,<sup>1</sup> Zhao Yonggang,<sup>1</sup> Wang Xianggao,<sup>2</sup>  
Shen Hongtao,<sup>3</sup> Wang Xiaoming,<sup>1</sup> Dou Liang,<sup>1</sup> Pang Fangfang,<sup>1</sup> Xu Yongning,<sup>1</sup> Zhao Qingzhang,<sup>1</sup>  
Yang Xuran,<sup>1</sup> Wu Shaoyong,<sup>1</sup> Li Kangning.<sup>1</sup>

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The determination of uranium isotopic composition in ultra-trace U samples is very important in different fields, especially for the nuclear forensics. A new measurement method with Accelerator Mass Spectrometry (AMS) technique has been developed for the analysis of uranium isotopic ratios in ultra-trace uranium samples at China Institute of Atomic Energy (CIAE), and as a result, the quality of about 5 nanogram level uranium samples analysis with AMS is achieved. Recently, the method was further optimized and developed by using series of different blank and standard samples. The results show that the quality of  $^{236}\text{U}$ -containing at the femtogram level in U-containing at nanogram level samples could already be analyzed by AMS technique at CIAE. The experimental setup, performances and results will be detailed in this contribution.

# **$^{129}\text{I}$ -AMS Analysis and Application Research in National Center of AMS (Xi'an) in Last 5 Years**

**Liu Qi<sup>1,2,3</sup> Zhou Weijian<sup>1,3</sup> Hou Xiaolin.<sup>1,3</sup>**

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[2]University of Chinese Academy of Sciences (China)

[3]Shaanxi Key Laboratory of Accelerator Mass Spectrometry Technology and Application, National Center of AMS (Xi'an),(China)

The  $^{129}\text{I}$  analysis and application research was started in 2009, two years after the opening of National Center of AMS (Xi'an). A series of chemical separation methods of iodine from different types of samples have been established including solvent extraction and combustion followed by extraction or co-precipitation depending on the sample types and the iodine content, the novel carrier-free method by  $\text{AgI-AgCl}$  co-precipitation was developed for ultra-low level iodine separation. The annual  $^{129}\text{I}$  sample preparation capability and AMS analysis capability has got to 800 samples and 1500 targets respectively, which occupied 20 percent of the machine time, about 1000 hours. The AMS background for  $^{129}\text{I}/^{127}\text{I}$  is  $1.5 \times 10^{-14}$ , and the chemical procedure background is about  $2 \sim 4 \times 10^{-13}$ , the routine batch measurement procedures for different kinds of targets had been established and was capable to analyze the targets with  $^{129}\text{I}/^{127}\text{I}$  ratio of  $2 \times 10^{-13} \sim 10^{-9}$ , the minimum iodine content in the target can be down to 5ug with  $^{129}\text{I}/^{127}\text{I} > 1 \times 10^{-12}$ . So far, more than 4000 targets had been measured for the environmental and geological application research using either naturally produced or anthropogenic  $^{129}\text{I}$ .

Topic : PRE 28

Poster Session 2

## The effect of AMS $^{13}\text{C}$ values on $^{14}\text{C}$ ages measured on a 0.5MV NEC compact accelerator

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Many AMS systems can measure  $^{14}\text{C}$ ,  $^{13}\text{C}$  and  $^{12}\text{C}$  simultaneously thus providing  $\delta^{13}\text{C}$  values which can be used for fractionation normalization without the need for offline  $^{13}\text{C}/^{12}\text{C}$  measurements on isotope ratio mass spectrometers (IRMS). However AMS  $\delta^{13}\text{C}$  values on our 0.5MV NEC Compact Accelerator often differ from IRMS values on the same material by 4-5‰ or more. It has been postulated that the AMS  $\delta^{13}\text{C}$  values account for the potential graphitization and machine induced fractionation, in addition to natural fractionation, but how much does this affect the  $^{14}\text{C}$  ages or  $\text{F}^{14}\text{C}$ ? We present an analysis of  $\text{F}^{14}\text{C}$  as a linear least squares fit with AMS  $\delta^{13}\text{C}$  results for several of our secondary standards. While there are samples for which there is an obvious correlation between AMS  $\delta^{13}\text{C}$  and  $\text{F}^{14}\text{C}$ , as quantified with the calculated probability of no correlation, we find that the trend lies within one standard deviation of the variance on our  $\text{F}^{14}\text{C}$  measurements. Our laboratory produces both zinc and hydrogen reduced graphite, and we present our results for each type. Additionally, we show the variance on our AMS  $\delta^{13}\text{C}$  measurements of our secondary standards.

Topic : PRE 29

Poster Session 2

**The ANU SSAMS, 7 years running****Fallon Stewart,<sup>1</sup> Wood R.<sup>1</sup> Fifield L.Keith.<sup>1</sup>**

[1]Australian National University (Australia)

It has been 7 years since The Australian National University received the NEC SSAMS. During this time we have analysed 13000 unknown radiocarbon samples. In the last year we have added an automated graphite preparation line coupled to an Elemental Analyzer/Carbonate device Isotope Ratio Mass Spectrometer. The IRMS preparation devices generate the CO<sub>2</sub>, 10% of which goes to the IRMS with the remaining 90% being captured in the automated graphite line. With this setup we obtain IRMS ( $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$  on organics and  $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$  on carbonates) on the same CO<sub>2</sub> that is then measured on the SSAMS. We have also switched to using helium as the stripper gas and have improved instrument transmission from 34% to 42%. Details of these changes and their influence on results will be discussed.



Topic : SP 12

Poster Session 2

**Biomedical Graphite and  $\text{CaF}_2$  Preparation and Measurement at PRIME Lab.****Jackson George,<sup>1</sup> Einstein Jane,<sup>1</sup> Kubley Tom,<sup>1</sup> Caffee Marc.<sup>1</sup>**

[1]Purdue University, (United States)

The biomedical program at PRIME Lab has prepared radiocarbon and  $^{41}\text{Ca}$  as tracers for a variety of applications. Over the last decade we have averaged several hundred  $^{14}\text{C}$  samples and several thousand  $^{41}\text{Ca}$  samples per year. Biomedical samples pose challenges that are relatively rare in the AMS community. We will discuss how to prepare and compensate for samples that have isotope ratios above the dynamic range of AMS, high interferences, and small samples sizes. The addition of carrier and the limits it places on the final reported precision will be examined. In the case of  $^{41}\text{Ca}$ , the trade off in the chromatography between yield and sample cleanliness will be analyzed. Finally, we have learned that care and precision in communication with collaborators is very important and some of the common problems created by misunderstandings of AMS will be discussed. As part of our routine procedure, we prepare secondary standards that have isotope ratios commonly encountered in our applications. We use material from the Joint Research Centre's Institute for Reference Materials and Measurement : IRMM-3701/4, 3701/5, and 3701/6 and a standard produced by PRIME Lab for  $^{41}\text{Ca}$ . We use International Atomic Energy Agency's IAEA C-3, IAEA C-7, IAEA C-8, and an  $\sim 12.5$  x modern oxalic acid standard supplied by CAMS for  $^{14}\text{C}$ . We will discuss our precision, reproducibility and the relative agreement between our measured and the reported values for these materials.

## Improving capabilities of surface exposure age dating within New Zealand.

Zondervan Albert,<sup>1</sup> Ditchburn Bob,<sup>1</sup> Norton Kevin,<sup>2</sup> Vandergoes Marcus,<sup>3</sup> Mcdonnell Randall,<sup>3</sup>  
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[4]Natural Hazards Division, GNZ Science, (New Zealand)

[5]Lamont-Doherty Earth Observatory, Columbia University, (United States)

For the most commonly used terrestrial cosmogenic nuclides,  $^{10}\text{Be}$  and  $^{26}\text{Al}$ , quartz is the target mineral of choice [1]. Alongside improvements of in situ production rate calibrations and of AMS measurement techniques, many labs continue to refine the technique to extract pure quartz from various rock types. New Zealand contains extensive areas of quartz-bearing rocks. In particular, the greywacke and schist lithologies of the Southern Alps have proved to be well-suited to surface exposure dating [e.g. 2], despite low abundance of sand-sized quartz grains. Victoria University of Wellington and GNS Science have started a collaboration to refine the analytical capabilities within New Zealand. At present, the focus is on establishing practical limits of key parameters for  $^{10}\text{Be}$ , such as minimum sample size and AMS detection limit, for different lithologies with low quartz concentration and/or small quartz grain size. We will present preliminary results from a replication study in which we sampled and analysed greywacke boulders from Last Glaciation moraines in the Southern Alps. These boulders have previously been surface exposure dated by the LDEO Cosmogenic Nuclide Group [2]. Part of our study aims to achieve a more efficient and safe implementation of the hot phosphoric acid (HPA) technique to separate quartz from other minerals [3,4]. We compare degrees of purity and recovery against those of the more widely employed etching method using hydrofluoric acid. Initial results show that the HPA method has a factor two higher quartz yields, while visual inspection suggests that quartz purities are similar to those obtained via HF etching.

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[3] N.A. Talvitie, 1951, Analytical Chemistry 23(4), p623-626.

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Topic : SP 14

Poster Session 2

**Multi-isotope analysis coupled to radiocarbon measurements.****Fallon Stewart,<sup>1</sup> Wood Rachel,<sup>1</sup> Sasaki Hideo,<sup>1</sup> Latimore Andrew.<sup>1</sup>**

[1]Australian National University (Australia)

2012 saw the completion of an automated graphite preparation system for AMS radiocarbon measurement at the Research School of Earth Sciences, ANU. The system consists of an Elemental Analyser (EA) and carbonate preparation device coupled to a Sercon 20-20 stable isotope ratio mass spectrometer (IRMS) and an in-house built automatic graphite preparation line. Organic samples are combusted in the EA, gases are purified and ~10% of the gas goes to a high precision measurement of  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ , the rest of the  $\text{CO}_2$  gas is trapped from the Helium stream cryogenically. The helium is pumped away using turbo pumps and the trapped  $\text{CO}_2$  is automatically transferred to an individual graphite reaction vessel for conversion to graphite using hydrogen and a temperature of  $570^\circ\text{C}$ . Carbonate samples are reacted in 5.9ml glass septa vials under a helium atmosphere,  $\text{CO}_2$  is purified and 10% of the  $\text{CO}_2$  enters the IRMS for a high precision  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  or  $\delta^{15}\text{N}$  measurement. With this system we process 20 samples a day and we can obtain (depending on the starting material) stable isotope ratios on the exact material that we obtain a radiocarbon measurement on using our Accelerator Mass Spectrometer. Specific scientific examples of the utility of these additional measurements will be shown. Specifically this has proved extremely useful for bone and coral.

Topic : SP 15

Poster Session 2

# AMS-<sup>14</sup>C Analysis of graphite from aerosol filters prepared in an automatized graphitization unit (age III).

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AMS-<sup>14</sup>C applications often require the analysis of small samples. Such is the case of atmospheric aerosols since frequently only a low mass is available. The ion beam physics group at the ETH, Zurich, has designed an automated graphitization equipment (AGE III) for the routine production of graphite for AMS analysis. In this system the organic sample is combusted in an elemental analyzer (EA) and the CO<sub>2</sub> produced is transferred to a graphitization unit where is converted to graphite through the reduction of hydrogen. In this study, we explored the potential use of the AGE III for preparation of small samples (down to 50 mg) using reference materials and blanks as well as aerosol filters that had been directly analyzed for radiocarbon content by AMS. The graphite samples prepared in the AGE III yield reproducible <sup>14</sup>C values for masses ranging from 50-300 mg. We also present a study case, where we analyzed the <sup>14</sup>C from atmospheric aerosols collected in Mexico City and a Cuernavaca (a smaller city nearby Mexico City) in order to compare the source apportionment of biomass and fossil fuel combustion.

Topic : SP 16

Poster Session 2

## Graphitization made easy : new streamlined and automated graphitization lines at the Lalonde AMS facility.

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The Lalonde AMS system was commissioned this year in the Advanced Research Complex at the University of Ottawa. The  $^{14}\text{C}$  preparation lab is designed for high throughput with newly designed, largely automated 10-port graphitization lines. The modular construction supports the vacuum line, cooling assembly, ovens, and touch screen controls. The stainless steel vacuum lines were orbitally welded to ensure smoother interior joints and hence less  $\text{CO}_2$  adherence and cross contamination. Sample  $\text{CO}_2$  is pre-measured on a separate gas cleaning line so the iron powder is weighed accordingly to avoid splitting the gas sample. The cooling assembly was designed to provide equal, uniform cooling for each reaction module to optimize water extraction during graphitization. Each cooling cup is filled with Syltherm to maximize heat transfer from the water trap and is cooled to  $< -45^\circ\text{C}$  by Syltherm circulating through a chiller connected to a copper coil in the cup. All operations on the line can be controlled on a touch screen monitor using a Labview program. Oven and water trap temperatures and pressures are continuously monitored and the ovens and valves are all operated under program control, thus facilitating automation. Safety controls programmed into the software prevent human error, which can lead to sample loss or pump flooding. All of these features, especially the automation, contribute to a streamlined design, making graphitization easy, safe, and user-friendly.

Topic : SP 17

Poster Session 2

## Report on the sample preparation methods performed at the Radiocarbon Laboratory of the Fluminense Federal University (LAC-UFF) in Brazil.

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[5]Laboratorio de Radioecologia (LARA), (Brazil)

In this work we report the sample preparation methods for radiocarbon Accelerator Mass Spectrometry measurements used at the Radiocarbon Laboratory of the Fluminense Federal University (LAC-UFF) in Brazil. The sample preparation laboratory was installed in 2009 and since 2012 a NEC single stage AMS system is in operation in the Physics Institute. The first tests with reference material have shown that isotopic fractionation in the graphitization step and in the accelerator could be an important issue for our samples (Anjos 2013). We changed our graphitization procedure to the Zn and TiH<sub>2</sub> method at lower temperatures following Xu et al. (2007) in order to enhance yield and quality of graphite. We now use sealed pyrex tubes and graphitization takes place at 520°C. The results are in good agreement with consensus values for reference materials and also for unknowns measured at different laboratories (Macario et al. 2013). Up to now we have regularly prepared samples of wood, charcoal, soil and carbonates. Other sample materials tests are under way. After regular pre-treatment we combust organic samples in sealed quartz tubes with CuO and Ag and we hydrolyze carbonate samples in phosphoric acid. We have now two more vacuum systems where we clean the gas samples with dry ice/ethanol and liquid nitrogen traps. Several tests for reducing and understanding sample preparation background were performed both with our SSAMS system and at the Keck-CCAMS Facility at University of California, Irvine, USA (KCCAMS/UCI).

Anjos RM et al. 2013. NIM B 294(0) :173-5.

Xu X et al. 2007. NIM B 259(1) :320-9.

Macario KD et al. 2013. Radiocarbon 55(2-3) :325-30.

Topic : SP 18

Poster Session 2

 **$^{14}\text{C}$  Contamination Testing Using Wet Chemical Oxidation and a Gas Ion Source.**

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The use of radiochemicals enriched with  $^{14}\text{C}$  can contaminate work areas used for natural abundance radiocarbon measurements. It is often difficult to know whether equipment, samples or facilities have been affected so, prior knowledge and testing can be invaluable to ensure the isotopic fidelity of measurements. The current AMS testing procedure for  $^{14}\text{C}$  involves swiping the area of interest with a quartz filter moistened with isopropanol. The filter is then dried and combusted in a sealed tube to produce  $\text{CO}_2$  that can be reduced to graphite and measured. Two problems that arise are that dedicated preparation equipment is required to prevent cross contamination with real samples and, that small samples can be difficult to prepare. We have developed a method using wet chemical oxidation and a gas ion source to overcome these issues. The procedure involves swiping the area with a moistened filter and placing it in a septa sealed Exetainer vial. After drying the filter, an aqueous solution of chemical oxidant (persulfate) is added and the vial is purged with helium. The vial is heated and the produced  $\text{CO}_2$  is then measured using the gas ion source of the MICADAS system at ETH Zürich. As little as  $5\text{ }\mu\text{g C}$  can be measured in 6 minutes to give a rapid and convenient way of screening for contamination. A minimum of equipment is used in the process so the possibility of cross contamination is minimized. We will outline the procedure and some results of this method, as well as further developments to increase the throughput of analysis.

Topic : SP 19

Poster Session 2

**CH<sub>4</sub> Headspace Extraction Method for <sup>14</sup>C-AMS Measurements.****Xu Xiaomei,<sup>1</sup> Elder Clayton,<sup>1</sup> Lehman Jennifer,<sup>1</sup> Pack Mary,<sup>1</sup> Czimczik Claudia.<sup>1</sup>**

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Methane (CH<sub>4</sub>) is a powerful greenhouse gas that plays important roles in atmospheric chemistry, including tropospheric ozone formation. However, the geographical distribution and interannual variability of individual CH<sub>4</sub> emission sources are poorly understood. Measurements of radiocarbon (<sup>14</sup>C) and stable C isotopic (<sup>13</sup>C) content of CH<sub>4</sub> are useful tools in determining its sources and pathways, and thus can improve our estimates of individual CH<sub>4</sub> emission sources and future CH<sub>4</sub> levels. We have developed a rapid and reliable headspace approach method to extract dissolved CH<sub>4</sub> and CO<sub>2</sub> gases from waters for <sup>14</sup>C analysis by AMS and stable C isotope analysis by IRMS. An evacuated gas canister as large as 2 l attached to a needle is used to extract headspace gases from a 1 l septa sealed water bottle. The gas canister is then filled to one atmospheric pressure with UHP N<sub>2</sub>, which serves as a carrier gas in the latter extraction. On a flow-through vacuum line, the headspace CH<sub>4</sub> is extracted, combusted and reduced to graphite (Pack et al. In review). Optimum extraction conditions, such as water/headspace ratio, shaking time and extraction efficiency are evaluated. Backgrounds, precision and accuracy are also determined using known <sup>14</sup>C CH<sub>4</sub> standard gases of both modern and <sup>14</sup>C free content. With an extraction efficiency of ~70%, the method can be applied to relatively low CH<sub>4</sub> concentration waters. For example, 1 l of water with 12 μM CH<sub>4</sub> would give ~0.1 mg C and allow for satisfactory AMS measurements. Dissolved CO<sub>2</sub> is isolated in the same process and can be measured for <sup>14</sup>C if desired.

Pack M.A. Xu X. Lupascu M. Kessler J.D. Czimczik C.I. (In review) A rapid method for preparing low-volume CH<sub>4</sub> and CO<sub>2</sub> gas samples for <sup>14</sup>C-AMS analysis. Organic Geochem.



Topic : SP 20

Poster Session 2

## State of the art and perspectives of the $^{14}\text{C}$ sample preparation lines after the first 9 years of operations at the CIRCE Centre (Italy).

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CIRCE (Centre for Isotopic Research on Cultural and Environmental heritage) Accelerator Mass Spectrometer was installed in early 2005 and started its operations by measuring radiocarbon ( $^{14}\text{C}$ ) isotopic ratios. Recently, after about 9 years of operation, the original sample preparation line based on the conventional sealed tube zinc reduction line feed by vacuum sealed tube combustion (black line) developed in late 2007 was paired with 1 brand new twin line (red line) and 1 Elemental Analyzer combustion interface feed zinc reduction line (blue line). This paper synthesizes the current state of the art of the  $^{14}\text{C}$  sample preparation laboratory after about a decade of operation and more than 6000 samples processed looking toward possible developments of the preparation lines in order to i) increase the range of samples typologies potentially analyzable and ii) reduce the required amount of sample for dating paying attention in guaranteeing a high sample throughput and an optimal measurement procedure characterization (e.g. quoted measurement uncertainties). Main results arising from the characterization of the newly introduced preparation lines performances in terms of induced background, accuracy, precision and isotopic discrimination will be showed looking toward the possibility of micro samples  $^{14}\text{C}$  dating.

Topic : SP 21

Poster Session 2

**Kinetics and isotope effect during graphitization for AMS in helium atmosphere.****Uslamin Evgeny,<sup>1</sup> Panov Vsevolod,<sup>1</sup> Sushentseva Natalia.<sup>1</sup>**

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AMS target preparation involves first the oxidation of carbon (in sample of interest) to CO<sub>2</sub> and second the graphitization. Reduction of CO<sub>2</sub> involves two steps. The first is reduction of CO<sub>2</sub> to CO. Each Zn and H<sub>2</sub> may be used as reducers. The second is disproportionation over Fe, Co, Ni powder catalyst. Studying of kinetics and isotopic fractionation is essential due to these factors having a great impact on observed data. Isotopic fractionation could be studying with AMS and <sup>13</sup>C measurements. Zn's been found to be usable as reducer. A zinc use allows perform graphitization in He atmosphere. Kinetics of CO<sub>2</sub> reduction and kinetic isotope effect influence has been studied. The temperature dependence of reaction was study. 520°C was shown as optimal temperature for performing this process. A process of CO<sub>2</sub> reduction over Zn at 520°C and pressure of 1 - 2 atm is a first-order reaction. Reaction rate constant  $k = (2.8 \pm 0.3) \cdot 10^{-4}$  1/sec. Therefore reaction of CO disproportionation over catalyst is rate limiting step of the whole process. For isotopic effect studying graphitization reaction has been performed under conditions below : T = 520 °C, p ~ 1 atm, Fe(325 mesh), C = 5/3. Then the reaction had been stopped and the  $\delta^{13}\text{C}$  have been analyzed using IRMS method. That's how the kinetic isotope effect has been discovered. 250 min of executing the reaction was enough for the isotope fractionation to become negligible. We preformed measurements for several sets of samples pre- and post-graphitizing. The average deviation for each of three sets (8 OXII, 30 Ceylon gr. 30 starch) was shown to be a less than 1 ‰. The difference in average  $\delta^{13}\text{C}$  values in pre- and post- graphitized samples is not greater than 1 ‰. Notably that  $\delta^{13}\text{C}$  value is decreasing after graphitization.

Topic : SP 22

Poster Session 2

**A non-vacuum graphitization method for AMS.**

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[2]NSF Arizona AMS Laboratory, (United States)

Accelerator mass spectrometry (AMS) radiocarbon measurements of organic samples require combustion to CO<sub>2</sub> before graphitization. A method using Elemental analyzer (EA) for combustion and dual inlet needle for trapping of CO<sub>2</sub> was described by Olsen (2007). We added a small amount of zinc dust and iron powder in septa-sealed vial prior trapping. The carrier gas was not pumped away. Septa-sealed vial with CO<sub>2</sub> and exceed of He was transferred into heater block (520 degrees, 4.5 hrs). 10 OX-II and 6 anthracites were prepared individually by this method. AMS measurements were made in Tucson (NSF-Arizona AMS Laboratory). The average value for OX-II is 135.2 pMC and average value for "dead" anthracite is 0.3 pMC. The cost for one sample using this simple technique is about 5, each operator can produce up to 16 samples for 8h day.

Topic : SP 23

Poster Session 2

 **$^{14}\text{C}$  age of collagen and bioapatite fraction of Late Pleistocene bison teeth from Alaska, USA.****Cherkinsky Alexander,<sup>1</sup> Glassburn Crystal,<sup>2</sup> Reuther Joshua.<sup>3</sup>**

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[2]Anthropology Department, University of Alaska, Fairbanks, (United States)

[3]University of Alaska Museum of North, Fairbanks, (United States)

The research addressed the stability of bioapatite and collagen fractions of AMS dated steppe bison teeth. During the study of Alaskan prehistoric bison mobility using differences in the strontium and oxygen isotope compositions of the 3rd molars, the AMS radiocarbon ages of 8 molars were determined on the fractions of collagen extracted from dentine and bioapatite extracted from enamel. The specimens are from the Lost Chicken Creek drainage in east-central Alaska, and were collected by the Bureau of Land Management in the 1980's after they were recovered from Quaternary sediments that were exposed by placer mining activities. Two different fluvial terraces are present at the collection site and the specimens in this study are from the lower and younger terrace, which formed during the late Pleistocene. The specimens were deposited along the terrace, and identified 9 distinct stratigraphic units which date to between >50,000 years BP and ~4,000 years BP. In this experiment we analyzed : 1) the bioapatite fraction extracted from tooth enamel and 2) collagen extracted from the root dentine. All studied samples were very well preserved and gave high yield of the fractions. The  $^{14}\text{C}$  age of the studied samples varied across age ranges between  $17,360 \pm 50$  and  $43,370 \pm 300$  non-calibrated years BP. Such wide range of ages allows us estimate the stability of each fraction in subarctic permafrost conditions.

Topic : SP 24

Poster Session 2

## Investigation of hydrocarbon molecular background for small samples radiocarbon dating via AMS

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Background ist always the limiting factor in small sample AMS radiocarbon dating, especially critical is the hydrocarbon molecular background. At the AMS laboratory in Erlangen several studies on the origin of hydrocarbons have been made in order to discover the spot of production. Therefore graphite samples, combusted and reduced in the usual way, have been analysed using different spectroscopic methods (IR, Raman and NMR) and thermogravimetry to identify hydrocarbon compounds. In addition to the usual reduction with iron powder different methods using Mg and  $\text{Fe}_2\text{O}_3$  have been tested. In this poster we will show the attempt to investigate the structure and point of production of the hydrocarbons and compare different reduction methods concerning the background.

Topic : SP 25

Poster Session 2

**Flushing graphite reactors with  $^{14}\text{C}$  free  $\text{CO}_2$  for AMS  $^{14}\text{C}$  analysis of very old samples (> 45,000 years BP).****De Rooij Marietta,<sup>1</sup> Van Der Plicht Johannes,<sup>1</sup> Meijer Harro.<sup>1</sup>**

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We tested the application of flushing the graphitization reactor with  $^{14}\text{C}$  free  $\text{CO}_2$  instead of  $\text{H}_2$  for AMS  $^{14}\text{C}$  analysis of very old samples (> 45,000 yrs BP). Due to the higher polarity of  $\text{CO}_2$ , the exchange between flushing gas molecules and the adsorbed molecules on the graphitization reactor walls will be enhanced. It resulted in lower and more reproducible background samples, which increased the precision in the  $^{14}\text{C}/^{12}\text{C}$  ratio of the batch mean background (from  $\pm 0.04$  to  $\pm 0.02$ ). According to the internationally agreed convention (Olsson, 1989), the radiocarbon age can be determined from the  $\delta^{13}\text{C}$ -normalized activity  $14\text{aN}$  when this activity is larger than two times its standard deviation ( $14\text{aN} > 2 \times \sigma [14\text{aN}]$ ). The flushing of the graphitization reactor with  $^{14}\text{C}$  free  $\text{CO}_2$ , also resulted in a reduced precision in the  $^{14}\text{C}/^{12}\text{C}$  ratio of the batch mean  $\text{HOxII}$  standard (from  $\pm 0.005 \times 10^{-12}$  to  $\pm 0.010 \times 10^{-12}$ ). However, for very old samples, the standard deviation in the  $\delta^{13}\text{C}$ -normalized activity depends entirely on the background variability. Therefore, the combined effect reduces  $\sigma[14\text{aN}]$  from  $\pm 0.04\%$  to  $\pm 0.02\%$  on average, for background samples. This corresponds to lowering the radiocarbon dating limit from  $\sim 57.000$  yrs BP to  $\sim 63.000$  yrs BP. Here we report the flushing of the graphitization reactor with  $^{14}\text{C}$  free  $\text{CO}_2$  instead of  $\text{H}_2$ , which is new to the best of our knowledge. It is generally applicable for AMS  $^{14}\text{C}$  laboratories that want to analyze very old samples close to the radiocarbon dating limit.

Topic : SP 26

Poster Session 2

## Determination of $^{14}\text{C}$ Pelagic Ocean Values through Atomic Bomb Radiocarbon Dating of Dolphin Teeth.

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The increase of environmental  $^{14}\text{C}$  caused by atomic bomb testing and the subsequent change in concentration as it is moved through the biosphere has made it possible to date things in the near past precisely. We will calibrate pelagic ocean  $^{14}\text{C}$  values using teeth from known age dolphins and then apply this calibration to whale tissues. Dolphins are a good proxy for larger whales as these animals have similar patterns of behavior (i.e. up and down in the water column) and dolphin teeth are more accessible than those of beaked and sperm whales. In general, unlike those of primates, marine mammal teeth consist of layers of dentin built up around the tooth pulp with a relatively thin outer layer and tip composed of enamel. Often, the age at the time of death of a marine mammal is estimated through the counting of dentin layers. Our plan is to develop a calibration curve from the teeth of 4 infant dolphins (1989-1998) and 2 adult dolphins that had tooth layers deposited in the 1960s or 1970s. For whole infant teeth and the tips of the adult teeth we will use a chemical procedure derived from Ambrose (1990), involving the crushing and chemical demineralizing of the teeth which should contain carbon from the same period in the animal's life throughout. Subsequently, a chemical procedure derived from Ubelaker et al. (2006) will be used which involves chemical demineralization, mechanical separation of layers, and organic material isolation from dentin layers of the adult teeth.

Topic : SP 27

Poster Session 2

**Quartz sample preparation and chemistry at PRIME Lab.****Chmiel Greg,<sup>1</sup> Clifton Thomas,<sup>1</sup> Granger Darryl,<sup>1</sup> Caffee Marc.<sup>1</sup>**

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Measurement of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  from quartz is the most common application of cosmogenic nuclides for geologic applications. There are many published chemistries for extracting Al and Be using column chromatography; however they often suffer limitations. High levels of impurities (e.g. Al, Fe, Ti, Ca) may overload columns while some methods have poor separation of Ti from Be or contamination due to sparingly soluble salts (e.g. Mg, Ni in oxalic acid), resulting in impure final products or poor yields. This is a particular problem at a high-throughput facility such as PRIME Lab where we process a variety of sample types and train a steady stream of novice visitors in sample preparation. We have therefore developed a robust, rapid, and streamlined procedure for the extraction of Al and Be from quartz. Quartz is separated from sediment or crushed rock using a combination of froth flotation, magnetic separation, heavy liquids, and selective dissolution, and then analyzed for trace elements by ICP-OES. Each sample is spiked and dissolved following standard procedures, fumed to dryness in  $\text{H}_2\text{SO}_4$ , and converted to chloride form. The key to our chemistry procedure is a  $\text{pH}>14$  precipitation that effectively removes most impurities (Fe, Ti, Mg, Mn, Ni, Ca) from Al and Be, which are then easily separated by ion chromatography in 0.4 M oxalic acid. Be is adsorbed on a 2 ml cation column while Al passes through and is collected on a 2 ml anion column stacked underneath. Be and Al are eluted from their separate columns as chlorides and precipitated as hydroxides. Be is dried as a nitrate while Al is dried as a chloride; both are flame-calcined, leading to a high purity fine oxide powder that is easily mixed with binder for analysis by AMS.



Topic : SP 28

Poster Session 2

## Solutions adopted at the LM<sup>14</sup>C in Saclay, France to avoid interferences during the graphitization process

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The ARTEMIS facility in Saclay, France, measures the radiocarbon content of 4500 samples a year for French organizations working in an array of fields, including environmental sciences, archaeology and hydrology. Our AMS system needs the samples to be turned into graphite targets. First, CO<sub>2</sub> is extracted from the sample and graphite is produced by hydrogen reduction of the CO<sub>2</sub> over iron powder. Nevertheless, iron can also react with other molecules like H<sub>2</sub>O, SO<sub>2</sub> or halogen compounds extracted from the sample and, in this case, the CO<sub>2</sub> reduction may be stopped (ones says that the iron is poisoned). This behavior is often observed for "dirty" samples, in general natural organic matters, coming from soils with high sulfur or halogen contents. Our study was focused on the influence of sulfur because this element is often suspected in the poisoning of iron. Different kind of samples which had encountered problems during the reduction step were selected and their sulfur and carbon contents were measured by elementary analysis. The idea was to find a sulfur content or a sulfur/carbon ratio above which the reduction becomes impossible. Our results have shown that sulfur is clearly not the only pollutant for reduction and that natural samples are not good candidates for this specific study. Even if the nature of the poison cannot be precisely determined, we have developed protocols to obtain "clean" CO<sub>2</sub>. We will present these protocols and will discuss of their impact on the <sup>14</sup>C result.

Topic : SP 29

Poster Session 2

**Quality management of AMS-radiocarbon measurements in Leibniz-Laboratory.****Huels Matthias,<sup>1</sup> Rakowski Andrzej,<sup>1,2</sup> Dreves Alexander,<sup>1</sup> Meadows John,<sup>1,3</sup> Schneider Ralph.<sup>1,4</sup>**

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[2]Institute of Physics - Center for Science and Education, (Poland)

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As required in any analytical measurement technique, a number of known samples had to be added to control the reliability of measurements of unknown samples. Over the past 3 years, frequent occurring measurement errors, e.g. apparently to old measured sample ages, failed to be detected by the existing quality management, consisting basically of the control of measurements done on primary standards, background samples, IAEA sample materials and recently added double measurements of unknown samples. Here we present results of our new quality control protocol, including the measurement of a number of known samples going through chemical sample treatment (tertiary standards), sealing and combustion (secondary standards), and primary standards (oxalic acid).

**Background correction for organic samples in Leibniz-Laboratory.****Huels Matthias,<sup>1</sup> Rakowski Andrzej<sup>1,2</sup> Nadeau Marie-Josee,<sup>3</sup> Grootes Pieter.<sup>4</sup>**

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The age limit of AMS radiocarbon dating is defined in principle by the detection limit. In addition to a machine background, infinitely old samples could a) contain an inherent contamination with  $^{14}\text{C}$ , and b) become contaminated with  $^{14}\text{C}$  during required sample preparation for AMS- $^{14}\text{C}$  measurements. As shown for carbonate samples, which have species-dependent  $^{14}\text{C}$  backgrounds (Nadeau et al 2001), we explore different organic materials such as wood, coal, and bone for material-specific apparent  $^{14}\text{C}$  background signatures. While samples such as coal or plant materials show rather comparable apparent background  $^{14}\text{C}$  concentrations, bone material contains a larger background signal. Whether differences in apparent background signals are related to differences in sample pretreatments, e.g. acid-base-acid vs. acid-base-acid plus gelatinization, or to a material-specific inherent  $^{14}\text{C}$  contamination, need further investigations. In any case, for old samples with  $^{14}\text{C}$  concentrations  $< 1\text{pMC}$  ( $\sim 37$  kyrs BP), a material-specific background correction can have large impacts on reported radiocarbon ages and may explain age discrepancies observed when comparing radiocarbon ages measured by different laboratories or comparisons to previously measured ages.

Nadeau M-J, Grootes P.M, Voelker A, Bruhn F, Duhr A, Oriwall A. 2001. Carbonate  $^{14}\text{C}$  background : Does it have multiple personalities ? Radiocarbon 43 (2A). p 169-176.

Topic : SP 31

Poster Session 2

**Successive searches of  $^{129}\text{I}$  contamination in the chemical sample preparation room.**

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We have addressed measurements of trace radionuclides such as  $^{36}\text{Cl}$  and  $^{129}\text{I}$  by AMS. It is important that the sample preparation room must be kept clean for accurate measurement. Clean means that it is far from not only visible state but also invisible contamination including isobars of interests. The contamination induces confusion in experimental results for AMS. However, the sample preparation room tends to be gradually contaminated in the process of chemical treating with a high concentration of radionuclides. In this work, we show the extent of contamination in the sample preparation rooms over time, particularly pertaining to  $^{129}\text{I}$ . Alkaline trap solutions were placed in the several sample preparation rooms with each three weeks. After the iodine carrier was added to the trap solutions, the trapped iodine was collected as  $\text{AgI}$  via chemical purification. Concentrations of  $^{129}\text{I}$  in the trap solutions were measured with the AMS at MALT, the University of Tokyo. In three month intervals, the  $^{129}\text{I}$  concentrations in the trap solutions were determined to be  $10^3$  to  $10^4$  atoms/g in the room where soil samples with high  $^{129}\text{I}$  level from Fukushima have been treated. On the other hand, we get results of  $^{129}\text{I}$  concentrations in trap solutions lower than  $10^3$  atoms/g in the room where we have only treated low  $^{129}\text{I}$  level samples. Besides, we find the results of highly  $^{129}\text{I}$  contamination ranging from  $10^5$  to  $10^6$  atoms/g in the room where we treated the neutron-activated iodine. The experimental results showed that levels of the  $^{129}\text{I}$  contaminations depend on ambient environmental conditions in the sample preparation room.

Topic : SP 32

Poster Session 2

**Radiochemistry of  $^{236}\text{U}$ ,  $^{129}\text{I}$  and Pu-isotopes in seawater samples.**

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Anthropogenic radionuclides are important tools in oceanography. In the last 3 years, several studies have applied  $^{236}\text{U}$  in the ocean and tested the prospective of this isotope to become a new oceanographic tracer. The combination of  $^{236}\text{U}$  in the ocean with other well-known AMS nuclides such as  $^{129}\text{I}$  and Pu-isotopes provides additional information and enables to trace water masses in the Arctic and Atlantic oceans. Therefore, setting up robust methods for the preparation of seawater samples for AMS measurements of these heavy ions is a fundamental step. In this work we present a sequential extraction method for  $^{129}\text{I}$ ,  $^{236}\text{U}$  and Pu-isotopes applied to seawater samples collected in the Arctic Ocean during the GEOTRACES cruises in 2011 and 2012. Briefly, 10-20 L samples were collected and stored in plastic cubitainers. In the lab, a 1 L aliquot of this sample was used for the further analysis of  $^{129}\text{I}$  based on the method by Michel et al. (2007). The rest of the sample was acidified and spiked with  $^{233}\text{U}$  and  $^{242}\text{Pu}$ . The sequential extraction of U and Pu-isotopes is done by pre-packed TEVA and UTEVA resin cartridges placed in a vacuum box system. Our automatized procedure allows the separation of U and Pu isotopes of from 12 samples in parallel. The newly set up preparation scheme fully exploits the analytical capabilities of the Compact ETH Zurich AMS system Tandy that is capable of analyzing I, U, and Pu isotopes at ultra trace levels in environmental samples.

Topic : SP 33

Poster Session 2

## Solid CO<sub>2</sub> adsorbent sample pretreatment by Elemental Analyser combustion for AMS biocarbon measurements

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Traditional AMS radiocarbon sample pretreatment methods are well known for their laborious nature. To combat this, Elemental Analyser sample combustion method has become the method of choice in Laboratory of Chronology for converting samples into CO<sub>2</sub>. By using the EA combustion the time spent in sample preparation is greatly reduced. However, until now, certain sample materials, namely the solid CO<sub>2</sub> adsorbents, have been challenging to combust. Solid CO<sub>2</sub> adsorbents are used to collect flue gas samples directly from power plants. By analysing the radiocarbon levels of flue gas samples it is possible to calculate the consumption ratio of bio/fossil fuels at a power plant. This method can therefore be used as a monitoring method i.e. in emissions trading scheme (ISO 13833).

Solid adsorbent samples were packed into tin cups in N<sub>2</sub> atmosphere and combusted in EA in 1050°C into CO<sub>2</sub>. Samples were collected cryogenically with liquid nitrogen and transferred into automated HASE sample graphitization line (Palonen et al., 2013). <sup>14</sup>C levels of the graphitized samples were measured at Helsinki AMS facility. In this presentation we will present the method, the pretreatment process and results and compare them to the standard (ISO 13833) acid treatment method.

ISO/FDIS 13833 :2012, Stationary source emissions - Determination of the ratio of biomass (biogenic) and fossil-derived carbon dioxide - Radiocarbon sampling and determination

Palonen, V., Pesonen, A., Herranen, T., Tikkanen, P. & Oinonen, M., 2013, HASE - The Helsinki adaptive sample preparation line. Nucl. Instr. Meth. Phys. Res. B : Beam Interactions with Materials and Atoms. 294, pp. 182-184

Topic : SP 34

Poster Session 2

 $^{129}\text{I}/^{127}\text{I}$  dating of Hokkaido underground fluids by AMS.

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The long-lived iodine isotope,  $^{129}\text{I}$  (half-life 15.7 My) is produced by the spallation of atmospheric Xe and by spontaneous fission of  $^{238}\text{U}$ . The ratio between  $^{129}\text{I}$  and the stable isotope  $^{127}\text{I}$  should be in steady state before being mixed with anthropogenic  $^{129}\text{I}$ . So this ratio was measured in order to provide an estimation of the age of iodine. In this study we analyzed  $^{129}\text{I}/^{127}\text{I}$  in underground waters with a high stable iodine concentrations in order to estimate the age of the dissolved iodine in sample collected from Hokkaido, northern island of Japan. It is known that several hot springs in Hokkaido contain high concentrations of halogens including iodine. However, the origin of the salts in these springs is not well known. We have collected hot spring waters from various places in Hokkaido. Iodine in the samples was separated by solvent extraction and it was precipitated as AgI to prepare a target for AMS. Analytical results showed the  $^{129}\text{I}/^{127}\text{I}$  ratios ranged between  $0.05 \times 10^{-12}$  and  $0.38 \times 10^{-12}$ . Samples collected from the north-western Hokkaido showed very low  $^{129}\text{I}/^{127}\text{I}$  ratios of  $0.05 \times 10^{-12}$  to  $0.1 \times 10^{-12}$ . Low values are observed along the longitude 141-142. These values are markedly lower than the  $^{129}\text{I}/^{127}\text{I}$  ratios observed in iodine-rich fluids in other areas in Japan, such as Chiba ( $0.18 \times 10^{-12}$ ), Niigata ( $0.3\text{-}0.4 \times 10^{-12}$ , Tomaru et al., 2009) and Satsuma-Iwojima ( $0.78 \times 10^{-12}$ , Snyder et al., 2002). Considering the  $^{129}\text{I}$  systematics (Fehn et al., 2004), iodine age in Hokkaido samples of the lowest  $^{129}\text{I}/^{127}\text{I}$  ratios is estimated to be 60-70 Ma. This indicates that the iodine-rich fluids are likely be derived from old marine sediment, which was later uplifted to form older rock formations in the present day coastal region of Hokkaido.

Topic : SP 35

Poster Session 2

 **$^{14}\text{C}$  determination in different bio-based products****Santos Javier,<sup>1</sup> Gómez-Martínez Isabel,<sup>1</sup> Agulló Lidia,<sup>1</sup> Reina María Teresa,<sup>1</sup> García-León Manuel.<sup>1,2</sup>**

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Radiocarbon determination can be used as a tool to investigate the presence of biological elements in different bio-based products, such as biodiesel blendings. Some of these products are liquid and thus the handling at the laboratory is not as straightforward as with solid samples. At CNA we have tested the viability of these samples using a graphitization system coupled to an elemental analyzer used for combustion of the samples. Specific equipment for liquid samples was tested. Measurement of samples was performed by low-energy AMS, paying special attention to background limits and reproducibility during sample preparation.



## Verification of cathodoluminescence application in process of mortars selection for radiocarbon dating

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Carbonatious mortars, taking into account the production process are the material possibly given the age of building construction. The real age of mortars is the age of binder, as the mortar is a mixture of binder and aggregate in different proportion. If the aggregate has the carbonatious character, the dead carbon effect could occur. It causes the overestimations of the results. It is connected with the presence of carbon partially or totally devoid of  $^{14}\text{C}$ . Another problem in mortars radiocarbon dating is connected with recrystallization causing the rejuvenation of the results. The usage of lime lumps is not well recognized yet in the light of radiocarbon dating. Depending on their origin, the lime lumps could give the real age of mortars or the rejuvenation of the  $^{14}\text{C}$  age. The environmental conditions of samples deposition is also an important issue in dating aspect. Many authors develop a method of samples preparation, basing on chemical (different fraction acid-leaching reaction) and mechanical separations (freezing and warming up) of mortars ingredients. The presented papers make an attempt to verify the application of cathodoluminescence analysis in samples selection for  $^{14}\text{C}$  dating. The experimental samples were prepared and together with archaeological mortars were observed and compared. Analysis were made on different kind of limestones, mortars with vary ingredients and carbonates of different age and origin.

**A simplified separation procedure for the preparation of Be and Al targets for AMS.****Binnie Steven,<sup>1</sup> Dunai J. Tibor,<sup>1</sup> Goral Tomasz,<sup>1</sup> Heinze Stefan,<sup>2</sup> Dewald Alfred.<sup>2</sup>**

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An extraction chromatographic resin (Beryllium resin) has been developed by Eichrom technologies to isolate beryllium from other metal ions. This resin has the potential produce high purity separates of beryllium and aluminum from dissolved quartz samples in a single column step, as opposed to the several stages more typically employed when processing AMS targets. The removal of interfering elements common in quartz such as titanium and iron, the retardation of the  $^{10}\text{Be}$  isobar boron, plus a complete separation of beryllium from aluminum are requisites for successful AMS measurements. Using these criteria we tested the Beryllium resin with the aim of simplifying AMS target preparation. It is apparent that previously published schemes using the resin to extract beryllium for environmental hazard monitoring are not appropriate for the milligram amounts of aluminum typical in the quartz masses dissolved for terrestrial cosmogenic nuclide studies. Consequently, we present an alternative protocol that shows the Beryllium resin allows for clean separations of beryllium and aluminum, both from each other and from potential AMS interferences. We further discuss the capacity of the resin for samples with high concentrations of interfering contaminants and compare  $^{10}\text{Be}$  and  $^{26}\text{Al}$  measurements on the Cologne-AMS from subsets of quartz samples prepared using both the Beryllium resin and more standard separation techniques.

Topic : SP 38

Poster Session 2

## Optimization of cosmogenic $^{10}\text{Be}$ and $^{26}\text{Al}$ extraction for precise AMS measurements of low concentrations.

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Both burial dating and recently introduced isochron-burial dating require accelerator mass spectrometry (AMS) analysis of low cosmogenic nuclide concentrations with low backgrounds and low uncertainties for more precise ages. Therefore, the aim of this study is to optimize the extraction of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  from quartz for the AMS measurements in the range of ca. 5000  $^{10}\text{Be}$  at.g<sup>-1</sup> and 50000  $^{26}\text{Al}$  at.g<sup>-1</sup> in 50 g of purified quartz. The latter is more feasible when total Al concentrations are ca. 20 ppm or less, with <10% uncertainty. To do this, we modified our sample preparation protocol. In order to reduce total Al concentrations, we added a treatment step with orthophosphoric acid. ICP-OES analysis of aliquots from different leaching steps yielded a dramatic decrease in total Al concentrations, e.g. from ca. 3000 down to ca. 10 ppm. To increase  $^{27}\text{Al}$  currents, Al was mixed with FeO, Cu, Nb, and Ti in two different molar mixing ratios (1:5 and 1:10). At the ETH Tandy AMS, the low energy current of two cathodes for each mixing ratio and metal was measured for >2000s. The best performance was found for Al plus Cu at a molar mixing ratio of 1:5.  $^{10}\text{Be}/^9\text{Be}$  and  $^{26}\text{Al}/^{27}\text{Al}$  measurements of several low concentration samples yielded ratios in the mid  $10^{-14}$  for Beryllium and low  $10^{-13}$  for Aluminum within less than 10% uncertainties, thus,  $^{10}\text{Be}$  and  $^{26}\text{Al}$  concentrations of ca.  $5000 \pm 500$  at.g<sup>-1</sup> and ca.  $50000 \pm 5000$  at.g<sup>-1</sup>, respectively. Additional tests are currently being carried out to further improve the performance of BeO samples containing very low amounts of  $^9\text{Be}$  carrier. The implications of our study are : (1) very young surface exposures, on the order of few hundred years, can be dated with few tens of years of uncertainty ; and (2) and isochron-burial dating of ca 100 ka old sediments is well possible.

Topic : SP 39

Poster Session 2

## Application of zinc reduction sealed tube graphitization on sub milligram samples using EnvironMICADAS

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The conventional graphitization procedures are designed to treat sample gas with 1 mg carbon content. However, in many cases, the amount of carbon extracted is much less than this volume. The demand for the radiocarbon analysis of samples containing less than 100  $\mu\text{g}$  carbon has increased over the past ten years. Such special samples may include human DNA, aerosol samples, crop residues, etc. Two approaches can be used to solve this problem. The use of a gas ion source is a good solution to analyze samples with 10-50  $\mu\text{g}$  carbon content using MICADAS type accelerator mass spectrometer. Our EnvironMICADAS is able to perform this kind of analysis routinely with moderate precision. The other solution is the development of a micro-graphitization method. The previously developed zinc reduction sealed tube graphitization is a good starting point for this process. The aim of this study was to find the appropriate parameters for the micro-graphitization of samples containing less than 100  $\mu\text{g}$  carbon. We analyzed how the graphitization time and the amount of zinc reagent and iron catalyst had influenced the  $^{12}\text{C}^+$  high energy ion current, transmission and the scale of the caused isotope fractionation in function of reduced carbon content of the samples. Another important task was to minimize the background level and increase the reproducibility of the graphitization process.  $^{14}\text{C}$  free  $\text{CO}_2$  gas, oxalic acid standard (NIST-SRM-4990c) and IAEA standards (C2, C5, C6, C8, C9) were used to perform the optimization steps. Finally, we have accomplished radiocarbon analysis of several real samples with the application of the developed micro-graphitization methodology.

This research was realized in the frames of TÁMOP 4.2.4.A/2-11-1-2012-0001.

Topic : SP 40

Poster Session 2

# **A new system for simultaneous IRMS and AMS radiocarbon measurements on gaseous samples : design features and performances of the gas handling interface.**

**Eugenia Braione,<sup>1</sup> Lucio Maruccio,<sup>1</sup> Gianluca Quarta,<sup>1</sup> Marisa D'Elia,<sup>1</sup> Lucio Calcagnile.<sup>1</sup>**

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We present the general design features and preliminary performances of a new system allowing the simultaneous AMS- $^{14}\text{C}$  and IRMS  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  measurements on samples with masses in the  $\mu\text{g}$  range. The system consists of an elemental analyzer (EA) , a gas splitting unit (GSU) , a IRMS system, a gas handling interface (GHI) and a sputtering ion source capable of accepting gaseous samples. The sample is first combusted in the EA which also performs the chromatographic separation of the combustion gases. The gas is then split into two fractions by the GSU, one is sent to the IRMS and the second to the GHI. The GHI is used to transport and inject the  $\text{CO}_2$  into the sputtering ion source. A movable LN trap is used to concentrate the  $\text{CO}_2$  in a reduced He flow which is then stored in a borosilicate gas-tight syringe connected via a glass capillary to a new gas ion source, installed in collaboration with the ETHZ Ion Beam Physics group. The control of the syringe piston by a stepper motor allows the injection of  $\text{CO}_2$  into the ion source with a constant and tuneable flow rate. The complete system is computer controlled and supports unattended operations.

**17.9 Friday 29 August - Morning****Topic : AHN 02****Session 13****Accelerator Mass Spectrometry applied to measure trace levels of actinides in underground detector experiments.**

**Galindo-Uribarri Alfredo<sup>1,2</sup> Batchelder Jon<sup>1,3</sup> Chu Ran<sup>1,2</sup> Fan Shiju<sup>1,2</sup> Juras Ray,<sup>1</sup> Liu Yuan,<sup>1</sup> Meigs Martha,<sup>1</sup> Mills Gerald,<sup>1</sup> Romero-Romero Elisa<sup>1,2</sup> Stracener Dan.<sup>1</sup>**

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We are developing new AMS techniques to measure impurity levels for materials used in underground detectors. In the search for neutrinoless double-beta decay in  $^{76}\text{Ge}$  an essential aspect is the production and use of ultra-clean Cu with extremely low levels of U and Th decay-chain contaminants. The AMS capabilities with the 25-MV Tandem at ORNL are being expanded. Methodology for measuring U and Th is being developed. An effort to increase the overall efficiency underway. This includes optimizing negative molecular ion production from a Cs sputtering source. A new high-intensity NEC Cs<sup>-</sup>sputtering ion source with a 40-position sample changer is being implemented. We are also pursuing a novel approach of producing with high efficiency positive ions followed by charge exchange [1]. Improvements on the detection systems such as a Bragg detector with digital signal readout and an ultra-thin SiN window are being developed. We have explored the use of negative ion Cu clusters to do a mass calibration of the injection magnet. This has led to a more general investigation of cluster formation. We have obtained mass-analyzed intensity spectra of large clusters of up to 50 atoms of C, Cu, and Au. We will present the first results and developmental activities to optimize the implementation of an AMS system to measure actinides.

Research sponsored by the LDRD Program at ORNL, managed by UT-Battelle, LLC, for the U.S. Department of Energy.

[1] Y. Liu, et al. this conference.

### A novel method for studying neutron-induced reactions on actinides.

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Improved and highly accurate nuclear data are urgently required for the design of advanced reactor concepts. This demand holds for minor actinides but also for the main fuel materials. Recent studies exhibit discrepancies at keV and MeV energies between major nuclear data libraries for neutron-induced reactions on  $^{232}\text{Th}$  as well as  $^{235}\text{U}$  and  $^{238}\text{U}$  that have great impact on the keff-value of fission reactors.

Neutron activation with subsequent AMS measurement of the reaction products represents an independent and complementary method to online particle detection techniques completely unaffected by any fission and other  $\gamma$ -ray background. Samples were activated with quasi-monoenergetic neutrons at KIT and IRMM for neutron capture studies with neutron energies between 25 keV and 5 MeV and fast neutron-induced reactions were studied in the energy range from 13 to 22 MeV. The reaction products were chemically separated and counted at four different AMS laboratories : VERA (3 MV tandem), ETH (0.5 MV Tandy), ANSTO (FN tandem) and ANU (14UD) taking advantage of highest sensitivities combined with interlaboratory comparisons for generating precise nuclear data.

Our results serve as important anchor points to solve present discrepancies in nuclear data libraries as well as impact on cross-section data used in the nuclear astrophysics community for s-process studies.

### Improved target preparation methods for actinides by AMS

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Ultra-sensitive detection of actinide isotopes is required in a number of important applications, including radiobioassay, nuclear forensics, nuclear waste characterization, environmental researches, biological tracer studies, and geochronology. Due to its excellent sensitivity, high rejection of interferences and low susceptibility to adverse sample matrices, accelerator mass spectrometry (AMS) has become the most sensitive, selective and robust technique to analyze the isotopic signature of long-lived actinides in biological and environmental samples.

The AMS ion sources require thermally and electrically conductive sample targets. The analyte isotopes and the isotopic tracers need to be homogeneously distributed in the AMS target to ensure accuracy and precision. The size of the target also needs to be minimized to obtain the highest signal intensity for good sensitivity. Despite this sub-optimal iron oxide targets continue to be extensively used for the determination of actinides by AMS, recently, new methods using mixed titanium/iron oxide and fluoride targets have been developed. To evaluate the method performance, samples spiked with femtogram levels of actinides (including Th, U, Np, Pu, Am, Cm and Cf isotopes) were prepared and measured using both compact Tandy system at ETH and the IsoTrace AMS. Improved detection limits of actinide isotopes in the sub femtogram range and good agreement between the measured and expected values have been achieved.



## The Use of Laser Ablation in an Electron Cyclotron Resonance Ion Source for Actinide Detection by AMS.

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Global research in Generation IV nuclear reactors and advanced fuel cycles has intensified the need for improved neutron capture cross section data of the actinides. The MANTRA (Measurement of Actinide Neutron TRAnsmutations) project will improve these data by measuring numerous energy-integrated cross sections across the actinide region. We will extract cross sections by measuring isotopic ratios from actinide samples, irradiated in the Advanced Test Reactor at INL, with Accelerator Mass Spectrometry (AMS) at ATLAS (ANL). In order to analyze the large number of samples needed for MANTRA and to meet the goal of extracting multiple cross sections per sample, we have made a number of modifications to the AMS setup at ATLAS. In particular, we are developing a technique to inject solid material into the ECR with laser ablation. With laser ablation, we can better control material injection and potentially increase efficiency in the ECR, thus creating less contamination in the source and reducing cross talk. In addition, we have installed an automated sample changer which allows us to quickly move between samples without altering source conditions. We will present our development work on laser ablation with the ECR, including a comparison of performance when using sputtering to ablate material. We will also present preliminary results of our first experiments with the multisample changer and laser ablation, and offer a future outlook for laser ablation in AMS experiments.

**Background reduction in  $^{236/238}\text{U}$  measurements.**

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The measurements of actinides isotopic ratios, in particular  $^{236}\text{U}/^{238}\text{U}$ , in environmental samples request high sensitivity. Moreover special effort has to be devoted to the suppression of interfering nuclides, such as  $^{235,238}\text{U}$ . At the AMS facility of CIRCE isotopic ratios down to  $\sim 10^{-10}$  are currently measured using a gas E- $\Delta$ E detector, while in order to push the limit towards natural levels a TOF-E system is used, featuring an MCP start detector and a Si stop detector. As the mass resolution of the latter is limited by the lay-out, an attempt to reduce the abundant isotope interference by other means has been undertaken. For U isotopic-ratio AMS measurements injection of  $\text{UO}^-$  is usually exploited, as justified by the good yield of this uranium molecular form in sputtering source. Nevertheless, it is known that a larger sputtering yield can be obtained by the  $\text{UC}_2^-$  molecule using cathodes containing a mixture of graphite powder and U. Another advantage of this choice is the absence of  $^{238}\text{U}^{14}\text{N}^-$  molecules injected with  $^{236}\text{U}^{16}\text{O}^-$ . Such cathodes, on the other hand, show a yield slowly increasing with sputtering time. In this study we report preliminary results on the characterization of the presence of molecular interferences comparing  $\text{UO}^-$ ,  $\text{UC}^-$  and  $\text{UC}_2^-$  cathodes, as well as on the possibility to stabilize the current yield from the source.

**Actinide Measurements by AMS and AS using Fluoride Matrices.**

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Actinides can be measured by alpha spectroscopy (AS), mass spectroscopy (ICPMS) or Accelerator Mass Spectrometry (AMS). We developed and tested a simple method to separate and then measure Pu and Am using single column extraction chromatography that allows the same samples to be measured easily by AS, ICPMS or AMS. Before loading the sample onto the extraction column the Pu was stabilized in the tetravalent oxidation state in concentrated HNO<sub>3</sub> with 0.05M NaNO<sub>2</sub>. Am (III) was adsorbed also onto the resin from concentrated HNO<sub>3</sub>, and desorbed with 0.1M HCl while keeping the Pu adsorbed. The on-column reduction of Pu (IV) to Pu (III) with 0.02M TiCl<sub>3</sub> facilitated the complete desorption of Pu. Interferences (e.g. Ca<sup>2+</sup>, Fe<sup>3+</sup>) were washed off from the resin bed with excess HNO<sub>3</sub>. Using NdF<sub>3</sub>, micro-precipitates of the separated isotopes were prepared for analysis by AS. These were subsequently dissolved and used for AMS measurement. Nd/Pu/F coprecipitates produced the strongest AMS beams of Pu and Am when they were diluted with about 7 to 10 :1 PbF<sub>2</sub>. The robustness of the AMS and AS measurements were validated using certified reference materials which agreed with the certified values over a range of about 1 to 100 Bq kg<sup>-1</sup>. The details of the AMS experimental procedures and the intercomparison results will be reported.

Topic : AHN 07

Session 14A

## Ultra-High Sensitivity Techniques for the Determination of $^3\text{He}/^4\text{He}$ Abundances in Helium by Accelerator Mass Spectrometry .

**Pardo Richard,<sup>1</sup> Hans Pieter Mumm,<sup>4</sup> Paul Michael,<sup>3</sup> Michael G. Huber,<sup>4</sup> Craig Huffer,<sup>5</sup> Paul Huffman,<sup>5</sup> C.L. Jiang,<sup>1</sup> Chris O'Shaughnessy,<sup>5</sup> Liang Yang,<sup>6</sup> Bauder William,<sup>1,2</sup> Collon Philippe,<sup>2</sup> Chithra Nair,<sup>1</sup> Tala Palchan,<sup>1</sup> Ernst Rehm,<sup>1</sup> Karl Schelhammer,<sup>5</sup> Robert Scott,<sup>1</sup> Richard Vondrasek,<sup>1</sup>**

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We report the development of an AMS technique to measure the  $^3\text{He}/^4\text{He}$  ratio using an RF discharge source and the ATLAS facility. Control over  $^3\text{He}/^4\text{He}$  ratio in helium are critical for experiments currently performed for re-determination of the neutron half-life. The ATLAS accelerator and beamline were tuned using  $^{12}\text{C}^{4+}$  ions produced in an Electron Cyclotron Resonance Ion Source (ECRIS). That configuration was then scaled first to  $\text{He}_3^{1+}$  ions from an RF discharge ion source coupled to the rear of the ECRIS, and then finally to the  $^3\text{He}^{1+}$  ions of interest produced also in the RF discharge source. The RF discharge source was developed for this experiment because of the large  $^3\text{He}$  background in the ECRIS plasma. The ions from the RF discharge source were transported through the (passive) ECR and accelerated ( $\sim 8$  MeV) through ATLAS.  $\text{H}_3^+$  and  $\text{DH}^+$

molecular ions are eliminated by dissociation through a gold stripper foil near the detector. The stripped ions were dispersed in a magnetic spectrograph and  $^3\text{H}^{2+}$  ions counted in the focal plane detector. This technique has been demonstrated to be sensitive to  $^3\text{He}/^4\text{He}$  ratios in the regime of  $10^{-12}$  with backgrounds that appear to be below  $10^{-14}$ . The techniques used to reduce the source backgrounds and remaining outstanding problems will be presented along with preliminary results from recent measurements on high purity  $^4\text{He}$  samples.

This work was supported by the US Department of Energy, Office of Nuclear Physics, under Contract No. DE-AC02-06CH11357.

Topic : AHN 01

Session 14A

**The AMS isotope Uranium-236 at VERA.**

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Over the last years, the Vienna Environmental Research Accelerator (VERA) was continuously extended to optimize the detection of the long-lived radioisotope uranium-236. It is now the first AMS system reaching the abundance sensitivity to address the expected typical natural isotopic ratios on the order  $^{236}\text{U}/^{238}\text{U} = 10^{-13}$ , while the improved detection limit of a few thousand  $^{236}\text{U}$  atoms significantly reduces the necessary size of anthropogenic samples. Stripping with helium to the 3+ charge state at 1.65 MV terminal voltage improved the yield by a factor of four, while a recently installed additional 90° magnet in the analyzer suppresses the background caused by  $^{235}\text{U}$  hydrides by several orders of magnitude. These developments allow measuring several hundred samples of  $^{236}\text{U}$  and other actinides per year in a comprehensive application program. Since  $^{236}\text{U}$  is ubiquitous in the environment, samples originate from freshwater, ocean water, corals, deep sea sediments, soil, peat, air filters, and the biosphere. The fields of applications are mainly environmental tracing, nuclear forensics, and radiation protection. First results on materials expected to be unaffected by anthropogenic  $^{236}\text{U}$  suggest that this contamination is more widespread than expected, and that improved chemical procedures have to be developed to fully exploit the instrumental limit. We will detail the VERA AMS system and present new results on  $^{236}\text{U}$  from the Fukushima exclusion zone as well as from a sediment-buried peat bog considered unaffected by anthropogenic influence.

**A new fast-cycling system for AMS at ANU.**

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AMS measurements using the 14UD Pelletron accelerator at the ANU are presently performed in a slow cycling mode whereby switching between isotopes is accomplished by changing the field in the mass-analysing magnet. Significant changes in ion source output limit the precision of the isotope-ratio measurements. In order to perform higher precision measurements, an upgrade of the ANU accelerator is underway. Fast switching times on the low energy side are achieved by holding the injector magnet field constant while changing the energy of the different isotopes by changing the pre-acceleration voltage after the ion source. First tests will be reported. At the high energy end a larger vacuum box in the analyzing magnet has been designed and is presently being manufactured and installed to allow the transport of differences in mass as large as 10% (e.g.  $^9\text{Be}$  and  $^{10}\text{Be}$ ) at constant terminal voltage. Currents of the stable beams (e.g.  $^9\text{Be}$ ) will be measured in offset Faraday cups after the analyzing magnet, and the appropriate vacuum housing and Faraday cups have been designed on the basis of detailed beam optics calculations using the code COSY Infinity [1]. For the cases where more than one isotope must be transported to the detector (e.g.  $^{239,240,242}\text{Pu}$  or  $^{233,236}\text{U}$ ) an additional refinement is necessary. If the accelerator voltage is to be kept constant, then the trajectories of the different isotopes around both the analyzing and switching magnets must be modified. This will be achieved using bunched electrostatic steerers before and after the magnets. Simulations have been performed with COSY Infinity to determine the optimal positions and sizes of these steerers.

[1] K. Makino et al. Nucl. Instr. Meth. Phys. Res. A 427 (1999) 338-343

Topic : AHN 08

Session 14A

**Reconstruction of anthropogenic  $^{236}\text{U}$  input to the Japan Sea.**

**Sakaguchi Aya,<sup>1</sup> Nomura Tomoya,<sup>1</sup> Steier Peter,<sup>2</sup> Watanabe Tsuyoshi,<sup>3</sup> Sasaki Keiichi,<sup>4</sup> Takahashi Yoshio,<sup>1</sup> Yamano Hiroya.<sup>5</sup>**

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$^{236}\text{U}$  ( $T_{1/2}=2.342\times 10^7$  y) has been used as a conservative tracer to clarify the oceanic circulation and the mechanism of deep water formation in the Japan Sea. However, the origin and amount of  $^{236}\text{U}$  input to the Japan Sea has not been clarified yet. In this study, we have focused on the analysis of coral, which retains the compositional information on surface seawater, in an attempt to reconstruct the detailed uranium isotopic composition ( $^{236}\text{U}/^{238}\text{U}$ ) of surface seawater of the Japan Sea. The coral sample (*Favia speciosa*) was retrieved using an underwater drill from a depth of 5 m at Kurosaki, Iki-island (N 33°48'22.5, E 129°40'02.9) in Nov/2012. This island is located in the Tsushima Strait which is the entrance of the dominant surface current of the Japan Sea, the Tsushima current. The total length of the coral core was 98 cm. The annual growth bands were identified by X-ray images and the variation of the Sr/Ca ratio obtained by LA-ICP-MS. The  $^{236}\text{U}/^{238}\text{U}$  in each annual ring was measured by AMS after appropriate sample preparation. Seventy eight annual growth rings were identified in 54 cm core indicating that the core represents the period from 1934 to 2012. The  $^{236}\text{U}/^{238}\text{U}$  atom ratios, which are a record of the ratio in seawater of the Tsushima Current, were in the range of  $4.51 \times 10^{-11}$  (1941) to  $6.15 \times 10^{-9}$  (1959). The highest ratio, in 1959, could be due to the maximum of hydrogen bomb tests conducted in 1958 at the Bikini and Eniwetok atolls on the North Equatorial Current, which is the origin of the Tsushima Current. As a consequence,  $^{236}\text{U}$  in the Japan Sea is dominantly derived from the Pacific Ocean as the Tsushima surface current rather than global fallout in 1963.

Topic : CNA 03

Session 14B

## Antarctica at the global "Last Glacial Maximum" - what can we learn from cosmogenic $^{10}\text{Be}$ and $^{26}\text{Al}$ exposure ages ?

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Ice volume changes at the coastal margins of Antarctica during the global LGM are uncertain. The little evidence available suggests that behaviour of the East and West Antarctic Ice Sheets are markedly different - and complex. It is hypothesised that during interglacials, thinning of the Ross Ice Shelf, a more open-water environment and increased precipitation, allowed outlet glaciers draining the Transantarctic Mnts and fed by interior Ice Sheets to advance during moist warmer periods, out of phase with colder arid periods. In contrast, glacier dynamics along the vast coastal perimeter of East Antarctica is strongly influenced by Southern Ocean conditions. Cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$  chronologies, although restricted to ice-free "oasis" and mountains flanking drainage glaciers, has become an invaluable, if not unique, tool to quantify Pleistocene ice sheet variability. Despite major advances, extracting reliable ages from glacial deposits in polar regions is problematic - recycling of previously exposed/ buried debris and continual post-depositional modification leads to age ambiguities for a coeval glacial landform. More importantly, cold-based ice advance can leave a landform unmodified resulting in young erratics deposited on "ancient" bedrock. Exposure ages from different localities throughout East Antarctica (Framnes Mnts, Lutzow-Holm Bay, Vestfold Hills) and West Antarctica (Denton Ranges, Hatherton Glacier, Shackleton Range) highlight some of the new findings. This talk presents results which quantify the magnitude and timing of paleo-ice sheet thickness changes, questions the validity of an "Antarctic LGM" and discusses the complexities presented by the geological spread observed in such studies



Topic : CNA 05

Session 14B

## An inherited cosmogenic burial signal from surface dune sands in the Simpson Desert dunefield, central Australia.

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Luminescence studies revealed complex dune activities within the extensive Australian dunefields during the late Quaternary. Initiation of these dunefields, however, dates beyond age-range of most stored luminescence signals and remains largely uncertain. Here we present a new study of cosmogenic burial dating applied to the Simpson Desert dunefield, central Australia. The simple burial ages, assuming a complete burial after deposition (i.e. no post-depositional nuclide production), calculated from  $^{10}\text{Be}$  and  $^{26}\text{Al}$  from 16 samples collected at 2-10 m depth in five dune sections, indicate 520-1860 ka. Eight samples with associated TL ages <15 ka, including three from dune surfaces, represent relatively uniform  $^{26}\text{Al}/^{10}\text{Be}$  ratios and thus simple burial ages with an average of  $660 \pm 70$  ka (S.D.). We interpret this as an "inherited burial signal" developed in sand particles prior to current dune formation. The origin of this apparent burial signal is uncertain. We hypothesise that it developed during particle residence time within the dunefield and/or during fluvial transport/storage prior to its addition to the dunefield. Nuclide inventory can then be modified only when the particle is isolated from surface mixing (i.e. by burial under a stable sediment column within dunes). Correcting this inherited signal (660 ka) for the deeper samples obtains inheritance-corrected burial ages of 230-1200 ka. The Simpson Desert, while still being actively reworked, appears to have developed over, at least, the last 1 Ma.

Topic : CNA 06

Session 14B

## New $^{10}\text{Be}$ evidence for Brunhes/Matuyama magnetic polarity reversal in Chinese Loess Plateau.

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Geomagnetic polarity reversals are generally considered to occur synchronously around the world, and are commonly used as time markers to correlate events between different sediment archives. However, the paleogeomagnetic studies have shown that the last geomagnetic polarity reversal separating the Brunhes and Matuyama chronozones in loess is found in glacial loess stage L8 (Zhou and Shackleton, 1999) which is correlated with Marine Isotope Stage 20 (MIS 20) (Lisiecki and Raymo, 2005), while in most marine sediment records, this reversal is found in interglacial sediments of MIS 19 (Tauxe et al. 1996; Zhou and Shackleton, 1999) at a position that is stratigraphically younger by at least  $\sim 25$  ka than that in Chinese loess, leading to the debate on uncertainties of paleoclimatic correlation between the Chinese loess-paleosol sequences and marine sediments (Wang et al. 2006; Liu et al. 2008; Jin and Liu, 2011). This asynchronicity has been attributed by some to post-depositional magnetic overprinting of loess, while others have argued that it is due to errors in the loess timescale. Here we solve this long-standing debate by exploiting a new method to extract reproducible records of geomagnetic field intensity from loess with  $^{10}\text{Be}$ -a proxy for global average geomagnetic field intensity-and use it to show that a pronounced minimum in field intensity-a requirement for dipole field reversal-is recorded in our loess records at ca.  $780 \pm 3$  ka BP. This timing is synchronous with the B/M reversal timing seen in marine records, verifying the standard loess timescale as correct, but it is  $\sim 25$  ka younger than the age of the magnetic polarity reversal recorded in these same Chinese loess sediments, demonstrating that loess magnetic overprinting has occurred.

## Combined cosmogenic $^{10}\text{Be}$ and $^{36}\text{Cl}$ nuclide concentrations constrain subglacial erosion rates.

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Concentrations of cosmogenic  $^{10}\text{Be}$  as well as  $^{36}\text{Cl}$  are frequently used by geomorphologists to calculate surface exposure ages of various landforms such as landslides or glacial moraines. For this purpose, it is commonly assumed that no cosmogenic nuclides were initially present in the rock, before the event to be dated. In the context of glacially formed landscapes, subglacial erosion of 2-3 meters of bedrock during the period of ice coverage suffices to remove any previously accumulated  $^{10}\text{Be}$ . In contrast, stronger contributions of muonic production pathways cause  $^{36}\text{Cl}$  to be continually produced at greater depth. Insufficient subglacial erosion leads to overestimation of surface exposure ages, if inherited nuclides are not corrected for. On the other side, a discrepancy between  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  concentrations carries information about the depth of bedrock removed during the Little Ice Age. Likewise, if the time since the retreat of the glacier is known independently, subglacial erosion depths can be determined based on the discordant concentrations of a single cosmogenic nuclide. We apply this multi-nuclide approach to several study sites in the Alps. Here we present data measured at the TANDY and TANDEM AMS facilities of the Laboratory of Ion Beam Physics at ETH Zürich. Interpretation of the data is aided by a MATLAB model simulating periods of exposure or glacial cover of user-definable length and erosion rates.

Topic : GAA 24

Session 14B

## A Bayesian approach to estimating in situ cosmogenic nuclide production rates that explicitly considers erosion.

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Production rates are a cornerstone in applications of in situ cosmogenic nuclides to surface exposure dating, erosion rate/denudation rate estimates, and burial dating. The most common approach for estimating production rates is to measure cosmogenic nuclide samples from sites with independently well-constrained exposure histories. Researchers attempt to minimize the effect of erosion through careful site and sample selection, such that its magnitude can either be considered negligible or can be constrained from differential relief between minerals with contrasting weathering properties such as quartz and feldspar. However, published calibration data for in situ cosmogenic  $^3\text{He}$  suggests surface erosion may be underestimated. In a number of instances, predicted sea level, high latitude (SLHL)  $^3\text{He}$  production rates tend to decrease with increasing surface age when minimal erosion is assumed. We address the difficulties in estimating the magnitude of long-term erosion on cosmogenic  $^3\text{He}$  production rate calibration by using Bayesian methods that incorporate a realistic range of outcrop erosion rates in the form of an exponential distribution, where higher erosion rates becomes less and less likely. Cosmogenic  $^3\text{He}$  provides an ideal test-bed for our approach, as it has the most calibration sites of the commonly measured cosmogenic nuclides, covering a broad spatial and temporal range. Results to date suggest that our approach largely reconciles previous discrepancies between sites of widely varying age, even at latitudes where geomagnetic effects are significant. In addition, we use Bayesian techniques to produce a best-estimate global SLHL  $^3\text{He}$  production rate that appropriately weights outliers without excluding them.

**17.10 Friday 29 August - Afternoon****Session 15****Topic : CRI 02****Session 15****Preparation of New Sets of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  AMS Standards****Kunihiko Nishiizumi.<sup>1</sup>**

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AMS measurements require normalization to primary standards; these standards are essential for all AMS measurements. Our  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ , and  $^{41}\text{Ca}$  AMS standards have been used as primary normalization standards at many AMS laboratories. Because of increasing demand of AMS standards, availability of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  standards that were prepared in 2001 were decreased. We have undertaken the task of making a new sets of AMS standards that should serve the community for the next  $\sim 20$  years. After consultation with the members of the AMS community we determined the target concentrations and ratios. In particular, the ratios of the new standards will differ from those prepared in 2001 to avoid potential mislabeling issues. We will also prepare a larger quantity of those standards that are most heavily utilized. We will conduct a rigorous inter-laboratory comparison of the standards before they are sent to users.

**$^{10}\text{Be}$**  : We found suitable a Be carrier after searching for more than 2 years. The  $^{10}\text{Be}/\text{Be}$  ratio in the carrier is  $5 \times 10^{-15}$ . Chemical impurities in the carrier are also very low, less than a few ppm of major elements. Six different concentrations of  $^{10}\text{Be}$  AMS standards were prepared from stock solution 2001-4 ( $1.082 \times 10^{-9}$ ) by sequential dilution of  $\sim 10$  kg of 36.6 mg Be/g carrier solution. The ratios of the new standards will range from  $1.0 \times 10^{-13}$  to  $2.5 \times 10^{-11}$ .

**$^{26}\text{Al}$**  : Six different concentrations of  $^{26}\text{Al}$  AMS standards were prepared from stock solution 2001-3 ( $1.880 \times 10^{-9}$ ) by dilution of a 35.7 mg Al/g carrier solution that was prepared from 188 g of high-pure Al metal. The ratios of the new standards will range from  $1.5 \times 10^{-13}$  to  $5.0 \times 10^{-11}$ .

I wish thank AMS colleagues at PRIME, CAMS, and MALT for initial test of preparation of new sets of AMS standards.

**Study on  $^{41}\text{Ca}$ -AMS technology for early diagnosis of cancer bone metastasis.**

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The annual incidence of new cancer patients in China is about 2 million, 50-60% of which will end up with bone metastasis. Profound study on the mechanism and early diagnosis of cancer bone metastasis are very significant for the prevention and treatment of bone metastasis, and the improvement of the survival rates for cancer patients. In order to monitor the processes of bone metabolism and early detection of bone metastasis of cancer cells, a technique of  $^{41}\text{Ca}$  isotope tracer combined with AMS has been developed and applied in the study on the bone metastasis of cancer cells by mouse simulation. In this work, 3-week-old female SD rats were randomly divided into five groups, each group were performed by injecting tumor cells into left upper thigh muscle, tail vein, femoral artery, femur, and knee joint, respectively, to establish the rat models for bone metastases. The most appropriate model ( thigh muscle group) was finally adopted in our real metastases experiment. Each rat in model group received an intramuscular injection of 250 $\mu\text{l}$   $\text{CaCl}_2$  Solution (containing 1.4 mg Ca and 5nCi  $^{41}\text{Ca}$ ) . One month later, the Walker 256 (rat mammary gland carcinoma cells) was injected into the model group with the established protocol. The Sequential urine and blood samples were collected and analyzed for total calcium and  $^{41}\text{Ca}$  content. The  $^{41}\text{Ca}$  and Ca in the Sequential urine and blood samples were measured by AMS and flame atomic absorption spectrometry (FAAS), respectively, after Microwave-Digestion. The longitudinal urinary  $^{41}\text{Ca}/\text{Ca}$  measurements may sensitively reveal skeletal perturbations, enabling improved clinical management through rapid identification of therapeutic success and non-invasive detection of the earliest stages of cancer growth in bone.

## Development of a Cs Isotope Measurement Technique for AMS.

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During nuclear weapons testing several caesium isotopes were released into the environment.  $^{137}\text{Cs}$  has been used in many studies. However this isotope has a relatively short half-life (30a) and it has already undergone  $\sim 50$  years of decay. Caesium 135, another fissile isotope, has a much longer half-life (about 2Ma) and could be used to replace  $^{137}\text{Cs}$  and the ratio between the two isotopes of Cs could be used to identify the source of Cs and to calculate the age of the source material. However  $^{135}\text{Cs}$  can be very difficult to measure. It cannot be gamma counted, as it is a pure beta emitter and beta counting is an impractical approach due to low decay rate. This leaves mass spectrometry as a viable option. Analyses using TIMS and ICP-MS have been established but their detection limits and sample preparation requirements suggest the possible use of AMS. The development of an AMS technique for  $^{135}\text{Cs}$  requires the development of (1) a beam of Cs anions, (2) a method to separate  $^{135}\text{Cs}$  from  $^{135}\text{Ba}$  and other ions with the same mass to charge ratio and (3) production of standards and yield tracers to measure the efficiency of the analytical process. We have used the IsoTrace AMS facility and : (1) Tested a number of different Cs compounds to identify methods to produce Cs beams, (2) Successfully separated  $^{135}\text{Cs}$  from  $^{135}\text{Ba}$  using an Isobar Separator for Anions (ISA). This reaction chamber selectively reacts  $^{135}\text{Ba}$  with oxygen while allowing  $^{135}\text{Cs}$  to pass into the accelerator and (3) Used  $^{134}\text{CS}$  as an internal standard and yield tracer. Currently, the limitations in the analysis of  $^{135}\text{Cs}$  are the beam current and cross contamination during sputtering. An array of different Cs molecules are being tested and optimized for greater and more stable beam currents.

# 18 List of registrants on June 11 2014

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